CHARACTERIZATION OF THE BACKGROUND NATURAL AEROSOL IN THE WEST OF IRELAND

FINAL TECHNICAL REPORT

S. G. JENNINGS

NOVEMBER 1988

European Research Office
United States Army Research, Development
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223-231 Marylebone Road
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The first coordinated joint field measurement program of aerosol size measurements at Mace Head and at Great Dun Fell was successfully carried out. The results showed that the aerosol levels are largely preserved - at least at the lower sizes of one particular size range - as the aerosol is transported in the prevailing SW air stream from Mace Head across to Great Dun Fell. Recommendations for further comparative work in this area are advocated. In addition the continuation of this successful initial aerosol data base at Mace Head is recommended.

ABSTRACT

Continuous data base measurements of ambient aerosol particle size distributions were made at Mace Head research field station strategically located in the west of Ireland to sense background maritime air from over the Atlantic ocean and continental air from the European mainland. Selected measurement periods from December 1987 through to June 1988 were used in the work. The measurements show two to three orders of magnitude reduction in aerosol number, mass and surface distribution for the prevailing SW or W wind directions as compared to European continental air sources from the SE or E directions. Two log-normal modes characterized the size distribution data. The smaller mode generally possessed geometric mean radii, r_g, of between 0.07 - 0.08 µm and geometric standard duration, σ_g, of about 1.3, whilst the larger mode was characterized by r_g, σ_g values of around 0.3 and 1.9. The lowest particle number and mass concentration values originated from sources from the westerly direction.

Electro-optical parameters of extinction coefficient (m^{-1}), σ_e, and backscatter coefficient (m^{-1} sr^{-1}), σ_b were calculated using the measured particle size distribution data at the Mace Head site. It was found that σ_e was generally insensitive to particle size (0.045 to 1.5 µm radius) for visible to near IR laser wavelength values and for middle IR wavelengths of 3.8 and 10.6 µm up to particle radius of 0.4 µm. In general, σ_b was independent of size for visible and near IR wavelengths. However for 3.8 and 10.6 µm laser wavelengths, σ_b increased by about four orders of magnitude for particle radius increase from 0.045 to 1.5 µm.

The first coordinated joint field measurement program of aerosol size measurements at Mace Head and at Great Dun Fell was successfully carried out. The results showed that the aerosol levels are largely preserved - at least at the lower sizes of one particular size range - as the aerosol is transported in the prevailing SW air stream from Mace Head across to Great Dun Fell. Recommendations for further comparative work in this area are advocated. In addition the continuation of this successful initial aerosol data base at Mace Head is recommended.

Keywords: geometric mean, atmospheric physics. (KR)

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CHARACTERIZATION OF THE BACKGROUND NATURAL AEROSOL IN THE WEST OF IRELAND

1. INTRODUCTION

Western Ireland is strategically positioned for natural aerosol background work, being the most westerly geographical location in Europe. The nearest major anthropogenic or man-made aerosol sources lie some three thousand miles west, along the eastern coast of the United States and Canada. The prevailing wind flow across Ireland (and Britain) is south-westerly in direction from the Atlantic ocean. This ensures that the natural atmospheric aerosol reaching western Ireland is a natural background aerosol, devoid of man-made or local influences.

The main proposed measurement programme described in the final report took place at the Atmospheric Physics field research station geographically located at probably the cleanest aerosol research site in western Europe, at Mace Head, Carna (approximately 53° 19'N, 9°51'W) in the west of Ireland about 55 miles west of Galway City. The location of the field station is strategically positioned and is exposed to a 130° sector (between 170° to 300° from N) in the prevailing SW-W wind from the Atlantic ocean. The site is essentially unencroached by land mass, being positioned a few hundred metres from the ocean. There is no human habitation along the prevailing wind direction between the site and the Atlantic ocean. Thus the field research station site is ideally located to measure the clean natural background aerosol, unpolluted by man made activity. Observations of size and concentration of Aitken nuclei have been made at the Mace Head site from 1958 to 1974 [O'Connor et al. (1958), O'Connor (1962), (1966)]. The site is distinctive in having very little change in its surroundings in over 25 years.

Studies of the physical characteristics, such as particle size or mass distribution of the natural background aerosol in the west of Ireland are very sparse. Aerosol studies to date have largely been carried out on Aitken nuclei. Information on aerosol mass loadings or optical characteristics of the aerosol are non-existent. A comparison of the change in aerosol properties and characteristics as the aerosol is advected from western Ireland across over Britain and onto mainland Europe had not been attempted up to now.

The main purpose of the proposed research programme was to characterize the natural background aerosol at the remote field research station at Mace Head in the west of Ireland. The characterization of the natural background aerosol as described here at this unique site establishes an important aerosol data base which can be extended in a possible later phase of the research programme.

The characterization of the natural aerosol is of crucial importance to the optical characterization of the atmosphere. Quantitative estimates of electromagnetic propagation of radiation requires knowledge of the size distribution, mass-loading and complex refractive index of the aerosol particles. Characterization of the atmospheric aerosol described in this Final Report allows the calculation of important optical parameters which form the second main objective of the research programme. Calculations are presented of optical characterization parameters such as extinction and backscatter coefficients together with lidar backscatter to extinction ratios--fundamental requirements in the operation and performance of E-O lidar systems for use in the remote sensing of the atmosphere. These calculations are based on the measured physical properties of the background aerosol measured on the site at Mace Head.

The work also involved collaboration with a parallel study conducted by John Latham, Department of Pure and Applied Physics, UMIST, Manchester, England together with his colleagues T.W. Choularton, M.W. Gallagher and K. Beswick. Coordination between the two research groups at Galway and Manchester permitted the performance of a simultaneous field aerosol case study at Mace Head and Great Dun Fell (a remote mountain field research station in northern England). This allowed the comparison of aerosol characteristics as the aerosol moved across western Ireland over the Irish and British land masses.

The final report describes the following:

(i) Characterization of aerosol particle number and size distribution for ambient air at the remote Atmospheric station site at Mace Head over the period December 1987 up to June 1988.

(ii) Aerosol mass loadings and Aitken nuclei concentration trends at Mace Head research station, coupled with typical Scanning electron microscope analyses of aerosol material present.

(iii) Collaborative field case study with the Atmospheric Physics Research Group under the direction of Professor John Latham, UMIST, Manchester, where comparisons are made between aerosol particle characterization parameters measured at Mace Head and at Great Dun Fell during suitable prevailing air trajectory conditions.

(iv) Detailed calculations of electro-optical parameters including extinction and backscatter coefficients, and lidar extinction to backscatter ratios based on the measured background aerosol parameters measured at

Mace Head, for radiation wavelengths from the visible to the middle-IR to include operational lidar wavelengths.

2. Experimental Apparatus and Techniques

The main instruementation used for particle size measuremnts was the Particle Measuring Systems (PMS) Active Scattering Aerosol Spectrometer Probe (ASASP-X). The instrument has the capability of measuring particle radius over the range 0.045 up to 1.5 micrometres (μm) over four ranges. The response characteristics of the probe have been investigated by Garvey and Pinnick (1983). Their results suggest that the manufacturer's instrument calibration is appropriate for spherical or slightly non-spherical particles having real refractive index $1.5 < n < 1.6$, so long as they are not highly absorbing (imaginary index < 0.005). In the work cited here, the probe was regularly calibrated for size response.

High volume sequential samples were used to measure the mass concentration of the total aerosol particulate loading. Use was also made of a compressor pump with a nuclepore filter head to permit scanning electronmicroscope (SEM) analyses of the aerosol. A Nolan-Pollak photoelectric nucleus counter was used to measure the ambient Aitken nucleus concentration. The meteorological parameters of wind direction and wind speed were measured using a Munro model IM 146 direction and velocity transmitter. Continuous recording of these important meteorological parameters was achieved via analogue signals accessed to a microcomputer.

3. Field Measurement Results

3.1 December 1987-March 1988

Field measurements of aerosol particle number and size distribution were carried out at Mace Head spanning the end of 1987 and up to June 1988 at about one monthly intervals. More frequent measurements were made of aerosol loading using a high volume sampler and of Aitken nuclei concentration using a photoelectric nucleus counter. A summary of the major field test dates over the contractual period are given in Table 1.

Table 1 Field Measurement Tests

Date	Prevailing Wind Direction	Comments
Dec.10-11,1987	Easterly	2 s data throughout. Solenoid valves in operation.
Dec.16-17,1987	Southerly	Some measurements bypassing solenoid valves were made.
Jan.13-15,1988	South-westerly-westerly	2s data
Feb.18-19,1988	-	Intercalibration of particle sizing instrumentation at UMIST, Manchester.
March 10-12,1988	Westerly	4 s data, no solenoid valves in use.
April 9-20,1988	Southeasterly to southwesterly	Parallel measurements at Mace Head and at Great Dun Fell.
April 20-22,1988	Southwesterly and easterly	Measurements at Mace Head
June 1-3, 1988	Southwesterly	4 s data throughout

Ambient Aerosol particle field measurements for all except the last two field tests listed in Table 1 were presented in the 3rd interim report.

Selected Data for the previous measurement periods are shown for the following periods: 11 December 1987 (Fig. 1); 17 December 1987 (Fig. 2); 14 January 1988 (Fig. 3); 10-11 March 1988 (Fig. 4). Aerosol concentration is presented using the format of particle concentration per cm^3 per micron which allows for weighting of the particle concentration according to particle channel width. Figures 1 to 4 show a decrease by over two orders of magnitude in aerosol particle concentration as the wind direction changes from an easterly direction (influx of continental European Aerosol) shown in Fig. 1, to a south-westerly direction (influx of unpolluted maritime air from across the Atlantic ocean), as shown in Fig. 3.

3.2 Period: April 19-22 1988

Atmospheric trajectories for the April 19-22, 1988 period are shown in Figs. 5 to 12. Trajectories were calculated for each day at 0Z and 12Z for the four pressure levels of 1000 mb, 850 mb, 700 mb and 500 mb.

The atmospheric trajectories were provided by Joyce Harris of The Geophysical Monitoring for Climatic Change (GMCC) Group within the National Oceanic and Atmospheric Administration (NOAA) at Boulder, CO. The trajectory program used is called "GAMBIT" for Gridded Atmospheric Multilevel Backward Isobaric Trajectories and is described by Harris (1982). One can see from Fig. 5 to 12 that with the exception of the 19 and 22 April, the Mace Head site was under a generally prevailing South-West air stream. Similar trajectory plots are available for the other field test dates. The measured data is presented here under the following formats:

(i) Aerosol particle concentration per cm^3 per micron as a function of particle radius.

(ii) Aerosol mass in grams per cm^3 per micron as a function of particle radius.

(iii) Aerosol surface area, cm^2 per cm^3 per micron as a function of particle radius.

(iv) Particle concentration plotted in the logarithmic format :dN/dlogr as a function of particle radius.

The data presented here is averaged over the selected period. Measurements were made every 4 seconds and recorded on data cartridges of 4.3 megabyte capacity via a microprocessor VGER operating system. The data was later transferred to the DEC 8700 mainframe computer and subsequently analysed.

Measured particle concentration, particle mass and particle surface distributions as a function of particle size are shown in Fig. 13 for the April 19th period, 1330-1600 local time. The mass is relatively constant throughout the size range suggesting that number concentration decreases as r^{-3} over this range. A log-normal fit to the data in the upper curve of Fig.14 yields two main modes. The log-normal mode parameter values of geometric mean radius, r_g and geometric standard deviation, σ_g are given in Table 2.

Fig. 14 also shows calculated values of extinction coefficient σ_e (m^{-1} μm^{-1}) and backscatter coefficient σ_b (m^{-1} sr^{-1}) based on the measurements of Fig. 13 as a function of particle radius for radiation wavelengths 0.55, 0.6934, 1.06, 3.8 and 10.6 μm. The Mie scattering algorithm of Dave (1968) was used in the Mie - Lorenz scattering code for E-O scattering calculations in this work.

The volume extinction and backscatter coefficients σ_e and σ_b of a polydispersion of spherical particles characterized by a size distribution n(r) and refractive index m are given by

$$\sigma_e = \int \pi r^2 Q_e n(r) dr, \qquad (1)$$

$$\sigma_b = \frac{1}{4\pi} \int \pi r^2 G n(r) dr, \qquad (2)$$

where $Q_e(m,x)$ is the Mie efficiency factor for extinction for a particle with refractive index m and size parameter $x = 2\pi r/\lambda$, and $G(m,x)$ is the backscatter gain defined as 4π times the ratio of the backscatter differential cross section to the geometric area.

Refractive index values were chosen to represent the more commonly found atmospheric constituents for maritime aerosol: sodium chloride and ammonium sulphate. For the purposes of calculation, two aerosol particle constituents were used. It was assumed that particles with radii > 0.4 µm (range 0 of the ASASP-X particle counter) consisted of sodium chloride and that all particles < 0.4 µm radius consisted of ammonium sulphate. This is a fairly good assumption based on separate volatility measurements carried out at the site. The change in extinction coefficient and backscatter coefficient with change in constituent (refractive index) is evident from Fig. 14. The extinction coefficient σ_e remains fairly uniform with particle size for the visible to near-IR wavelengths. There is a gradual increase in σ_e with radius > 0.4 µm in the middle IR.

Similar output plots as described above are shown in Fig. 15, Fig. 16, Fig. 17 and Fig. 18 for the following selected time intervals in the April data: April 20 : 1800-2400 and April 21 : 0150 - 1245. These periods are characterized by relatively constant levels of ambient particle concentration. The log-normal mode parameters of geometric mean radius r_g and of

geometric standard deviation σ_g are summarized in Table 2. The averaged values of particle number concentration, mass concentration and surface area concentration broadly exhibit similar characteristics and values for the three chosen periods in the April data. This is also borne out by the fitted log-normal parameters values of r_g and σ_g shown in Table 2.

Table 2 Log-normal mode Parameters for Aerosol size distributions at Mace Head

Geometric mean Radius, r_g (μm)		Geometric Standard Deviation, σ_g		Measurement Period
Mode 1	Mode 2	Mode 1	Mode 2	
0.083	0.22	1.25	2.35	April 19, 1988 1330 - 1600
0.076	0.29	1.44	1.91	April 20, 1988 1800 - 2400
0.092	0.30	1.25	2.15	April 21, 1988 0150 - 1245
0.072	0.30	1.32	1.88	June 1, 1988 1850 - 2150
0.077	0.30	1.34	1.89	June 2, 1988 0050 - 1000
0.074	0.30	1.30	1.92	June 2, 1988 1600 - 2000

3.3 Period: June 1 - June 3 1988

The measured data was quite uniform throughout the June period. A similar output format as for the April data is used here. Averaged output data is shown for June 1, 1850-2150 in Fig. 19 and Fig. 20; for June 2, 0050-1000, in Fig. 21 and Fig. 22, and for June 2, 1600-2000, in Fig. 23 and Fig. 24. Values of fitted log-normal parameters r_g and σ_g to the data are given in Table 2. The particle concentration distribution levels for number, mass and surface are similar for the first two periods in June when the wind is from about the 210° direction. As the wind becomes more southwesterly in direction, there is a reduction in particle number concentration throughout the particle size range. The wind speed remained relatively constant throughout these periods at about 8 ms^{-1} (or about 17 knots). The relatively uniform measured values of the aerosol particle distribution for the June period is reflected by the near agreement of the fitted log-normal parameters to the data as shown in Table 2.

The values of the extinction coefficient and backscatter coefficient curves shown in Fig. 20 and Fig. 22 of the first two periods in June are close in agreement. Both extinction and backscatter coefficients are reduced for the final June period with the wind in the SW direction, due to an overall reduction in aerosol particle concentration values associated with that particular direction.

The diurnal variation in aerosol particle concentration throughout all of the June period from 1850 on June 1st to 1550 on June 3rd is shown in Fig. 25. Range 3,2,1 and 0 are shown from the top curve downwards to the lowest curve in the Figure. The range in particle diameter corresponding to Ranges 3,2,1 and 0 is shown in Table 3.

Table 3 <u>Size calibration of Particle Measuring Systems (PMS) ASASP-X light scattering counter.</u>

	Range 3	Range 2	Range 1	Range 0
Particle diameter intervals (μm)	0.09-0.20	0.15-0.30	0.24-0.84	0.6-3.0

There is a general decrease in aerosol particle concentration with time, corresponding to a gradual change in wind direction which was initially at about 210°, then changed to SW for the period 24 to about 40 hours (0000 to 1600 hours on June 2nd), and then to a more westerly direction from 40 to about 59 1/2 hours (1600 hours on June 2nd through to 1130 on June 3rd). The abrupt dip in concentration at about 60 hours is due to a sudden change in wind direction from W to 300° and a change in wind speed from about 15 to about 25-30 knots (from about 27-30 km hr^{-1} to about 50 km hr^{-1}).

4. <u>Extinction to Lidar Backscatter Ratio</u>

The extinction to backscatter ratio is calculated for each of the particle probe size channels at wavelengths 1.55, 0.6934, 1.06, 3.8 and 10.6 μm. They are plotted in Fig. 26 and Fig. 27 for constituents ammonium sulphate and sodium chloride. It is seen that there is wide variation in the ratio with particle size for both constituents with the exception of the calculations at wavelength λ = 10.6 μm for NaCl, when the ratio is approximately equal to 10. The extinction to backscatter ratio is calculated for an aerosol particle size destribution with the smaller particle size mode consisting of ammonium sulphate possessing size values over range 3, 2 and 1 of the ASASP-X probe (see Table 3). It was assumed that the upper size range 0 consisted of sodium chloride. This partitioning is broadly

consistent with experimental findings from volatility measurements made at Mace Head. Fig. 28 demonstrates the result for the combined constituents of ammonium and sodium chloride of the extinction to backscatter ratio as a function of particle size. It is seen that the influence of refractive index on the lidar backscatter to extinction ratio is most sensitive for the 10.6 μm E-O wavelength.

5 Aerosol mass loading and Aitken nucleus concentration measurements

5.1 Aitken Nucleus Concentration Measurements

The total Aitken nucleus concentration, Z, was measured with a Nolan-Pollak photoelectric nucleus counter. The construction, calibration and performance of the counter has been described by Metnieks and Pollak (1959). Measurements of Aitken nuclei were normally made at weekly or two-weekly observations over the contractual period. The variation of nucleus concentration, Z, cm^{-3} with wind direction is summarised in Table 4 below.

Table 4 Variation of Aitken Nucleus Concentration with wind direction at Mace Head

Wind Direction	E	ESE	SE	S	SW	W	NW
Aitken Nucleus Concentration cm^{-3}	3733	3662	40323	572	431	222	634
Number of Observations	82	14	10	40	82	72	58

The tabulated results indicate that the cleanest air is coming from the W direction.

The variation of nucleus concentration as a function of time (shown in minutes) is given in Fig. 29 for the June 1 through June 3 period. The data gaps coincide with middle of the night episodes. Nucleus concentration of between 300 -700 per cm^{-3} are seen to be typical values for the prevailing SW wind for most of the period. The reduction in nucleus concentration at about the 2200 minute time interval of Fig. 29 corresponds to a change in wind direction to a westerly direction. The subsequent concentration increase is associated with the change in wind direction to

the 300° sector which is identified with an incursion of air from overland anthropogenic sources.

5.2 Aerosol Mass Concentrations

The aerosol mass concentrations ($\mu g\ m^{-3}$) has been measured at Mace Head Field Station on a regular basis. A filtered air hood has been installed at the site (as reported in the 2nd interim report) to facilitate clean handling of the Whatman 41 filters used in the Hi-vol sampling. The sampling protocol involves preweighing the filter which is first placed in a desiccator until a constant mass is obtained. The filter is then loaded into the high volume sampler and sampling normally takes place over a 1-2 day period. The total volume of aerosol sampled is measured. The exposed filter is then removed and placed in the desiccator until it yields a steady mass reading. Some 30 high volume particulate samples have been measured. An attempt to categorize the particulate mass according to wind direction sectors is shown in Table 5. This necessarily constitutes a smaller sample group since overlap in wind direction sectors sometimes occurred over a few days sampling period.

Table 5 High Volume Aerosol Particulate Mass Loadings

Wind Direction (Degrees)	Mass Concentration, $\mu g\ m^{-3}$			
0 - 90	26.0	29.8	16.6	
90 - 180	36.8	29.7	48.7	
180 - 270	17.0	36.1	32.0	28.1
270 - 360	10.5	29.7	23.0	24.9

More measurements are required in the future to draw statistically significant conclusions from the particulate mass data, although these measurements tend to indicate a higher mass loading from eastern directions from which continental air masses originate.

5.3 Nuclepore Filtration Sampling

A series of nuclepore filters of pore size 0.2 μm were used to sample the ambient aerosol at Mace Head. The aspiration pump was generally run for about twelve hours duration. The filters were subsequently analysed using the Scanning Electron Microscope (Hitachi S-570) facility at University College Galway. Typical scanning electromicrographs of aerosol particle samples for April 20, 1988 and April 22, 1988 are shown in Fig. 30. The upper picture for April 20 shows the dominant presence of salt particles (from the prevailing SW wind direction over the ocean) as evidenced from the characteristic crystal shape. In contrast, the lower picture shows a typical representation of aerosol transported over land from the Easterly direction for April 22. The density of particles is also clearly greater for the anthropogenic aerosol sources. Scanning electronmicrographs for the June 2 - 3 period with wind from the prevailing SW direction are presented in Fig. 31 and they show the characteristic shape of Sodium Chloride particles. The lower picture indicates a sizeable proportion of the particles with size greater than the upper sizing capabililty of the ASASP-X. X-ray energy dispersive analysis of the particles confirms the dominance of sodium and chlorine as evident in Fig. 32.

6 Collaborative Field Case Study with the Atmospheric Physics Research Group, UMIST, Manchester.

Initial collaboration took place between the Atmospheric Physics Group at University College Galway and UMIST, Manchester with a series of intercomparison tests performed between the Galway ASASP-X probe and the UMIST ASASP probe. Laboratory generated polydispersions of ammonium sulphate were used as the test aerosol. It was suggested (Beswick, private communication) that a future intercomparison test might also use the ambient outside aerosol as the test aerosol. Since the raw data was stored on computer at UMIST, it was agreed during the intercomparison tests that the intercomparison probe analysis would be carried out by the Atmospheric Physics Research Group (APRG) at UMIST. The results of the analysis showing the difference in particle concentration as a function of particle size for the two probes were evaluated (Beswick, private communication) and used to renormalize UMIST's ASASP concentration values, adopting Galway's ASASP-X probe as the standard probe. The ASASP-X probe is considered to give a true concentration reading as it is totally plumbed to measure particle concentration absolutely. The probe intercomparison data showed highly variable correction factors, both from range to range and from channel to channel, particularly for the lower channel numbers.

The period when comparison was possible between aerosol particle concentration measured at Mace Head and at Great Dun Fell was during the Spring timeframe during which time the APRG, UMIST were making multi-site measurements at Great Dun Fell involving at least two separate projects involving collaboration with separate research institutes (Gallagher, private communication).

From the beginning of UMIST measurement period on 28 March up to and including 19 April 1988, there was no favourable SW air flow from Mace Head to Great Dun Fell on the days of 28, 29 of March; 5,6,12,13,14, and 19 April (Gallagher, private communication) when UMIST's ASASP's were recording data; with the exception of about a 10 hour period [0100 -1100 local time at Mace Head] - on March 28th. It is not easy to predict well in advance favourable time slots for intercomparison purposes and the problem is best overcome (in hindsight) by recording continuously at both sites.

Favourable south west air flow conditions as forecasted existed at Mace Head for the period April 20 and April 21 (as evidenced in Fig. 7 to Fig. 10.). Field measurements at Mace Head were carried out over the 19 to 22 April period, which was communicated to the UMIST group at the time.

Particle size ASASP measurements were recorded at the Silverband site (650 m m.s.l.) on 20 April 1988 under high humidity conditions (from 86.5% to 100% relative humidity). The measurements were made using Range 1 of the ASASP-probe (range 0.2 to 0.5 μm radius). After excluding data with very high relative humidity (> 98%) the particle concentration per cc per second was plotted as a function of particle radius. Using a transit time of between about 10 to 12 hours between Mace Head and Great Dun Fell, averaged particle concentration measured at Mace Head is compared to the Great Dun Fell measurements in Fig. 33 and Fig. 34 for the two periods of 12.47.35 - 13.02.36 GMT and 13.02.36 - 13.17.36 GMT (corresponding to the two lowest relative humidity conditions of 87.5% and 86.5%). There is reasonably close agreement in particle concentration for the lowest 7-8 channels for the two sites. There is near complete particle attrition in the larger sized particle categories measured at Great Dun Fell.

The Great Dun Fell Silverband site is not ideal as a comparison site with Mace Head for sampling <u>dry</u> aerosol under conditions of high humidity as was the case for the 20 April data. The effect of humidity on dry particle growth would need to be taken into account in the comparison of dry aerosol particle number concentration at the two sites. In addition, aerosol loss with height (Mace Head is at about 17 m msl compared to Great Dun Fell at 847 m msl) would need to be accounted for in comparing values at the two sites.

It was not possible to compare additional continuous particle size distribution data taken at Mace Head on 20 and 21 April with the site at Great Dun Fell because the allotted time of that project at Great Dun Fell had terminated (Gallagher, private communication).

7. SUMMARY OF RESULTS

An aerosol particle sizing instrumentation system with continuous measurement capability (with data handling capability down to 2 Hz) has been successfully operated at Mace Head field research station over the period December 1987 through June 1988. The large volume of particle size data was transferred to and analysed by a DEC 8700 mainframe computer. Field test measuremnts covering the above period are presented of aerosol particle size, mass and surface distributions in the report. The measurements show between two or three orders reduction in magnitude of the above aerosol parameters when the wind is from a W or prevailing SW direction (that is for a maritime air mass) as compared to when the aerosol is of continental European origin from a predominantly E or SE direction. Curve fitting of the particle size data for the prevailing SW airmass at Mace Head indicated that in general there were two log-normal modes present. The smaller mode possessed mean radius of between 0.07 - 0.08 μm and geometric standard deviation of about 1.30 whilst the second mode was characterized by a geometric mean radius of about 0.3 μm and geometric standard deviation of about 1.9. Aerosol mass loadings varied from 10.5 μg m^{-3} (for W direction air trajectory) up to 48.7 μg m^{-3} for the E - SE wind sector of anthropogenic source. The mass measurements were complemented by Aitken Nucleus measurements at the site which showed that the lowest particle number concentration counts (220 cm^{-3}) originated from the W direction whilst the largest particle concentration (40,000 cm^{-3}) were from the SE direction.

It is considered desirable to continue with this initial data base measurement program at Mace Head.

The electro-optical parameters of extinction coefficient (m^{-1}) and backscatter coefficient (m^{-1} sr^{-1}) were calculated using a Dave Mie scattering code, but modified to yield reduced execution time. The scattering parameters were determined using the measured particle distributions at Mace Head. Particle constituents of ammonium sulphate and sodium chloride (based on separate volatility work) were used for the calculations. Extinction coefficient was fairly insensitive to the particle size range (0.045 to 1.5 μm radius) for the visible through to near IR values (and for 3.8 and 10.6 μm wavelengths for particle radius < 0.4 μm). There was an increase of about an order of magnitude in extinction coefficient from particle radius of 0.4 through to 1.5 μm radius over the wavelength range 3.8 to 10.6 μm.

The backscatter coefficient was broadly independent of particle size for visible through to near-IR wavelengths. For wavelengths of 3.8 and 10.6 μm, the backscatter coefficient increased by about four orders of magnitude for an increase in particle radius from 0.045 to 1.5 μm. Extinction to backscatter ratio showed variations up to two orders of magnitude with particle size with the exception of wavelength 10.6 μm for the sodium chloride constituent which yielded a constant value of about 8-10 for the extinction to backscatter ratio.

The first ever joint measurement programme of aerosol particle size distribution measurements made in a coordinated manner at Mace Head and Great Dun Fell research field station was carried out. The comparison of particle concentration data showed that the aerosol levels at the smaller size values are largely preserved as the aerosol is transported in the prevailing SW airstream from Mace Head across to Great Dun Fell. However, more comparison work is needed to be done which should include :

(i) The use of a larger particle size range - particularly towards the smaller radius end of the size spectrum.

(ii) measurements at approximately the same altitude and under similar meteorological conditions of temperature and relative humidity.

(iii) the use of continuous data at both sites.

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9. Acknowledgements

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The Atmospheric air trajectories have been provided by NOAA through the work of Joyce Harris of the Geophysical Monitoring for Climatic Change (GMCC) group in Boulder, CO, USA.

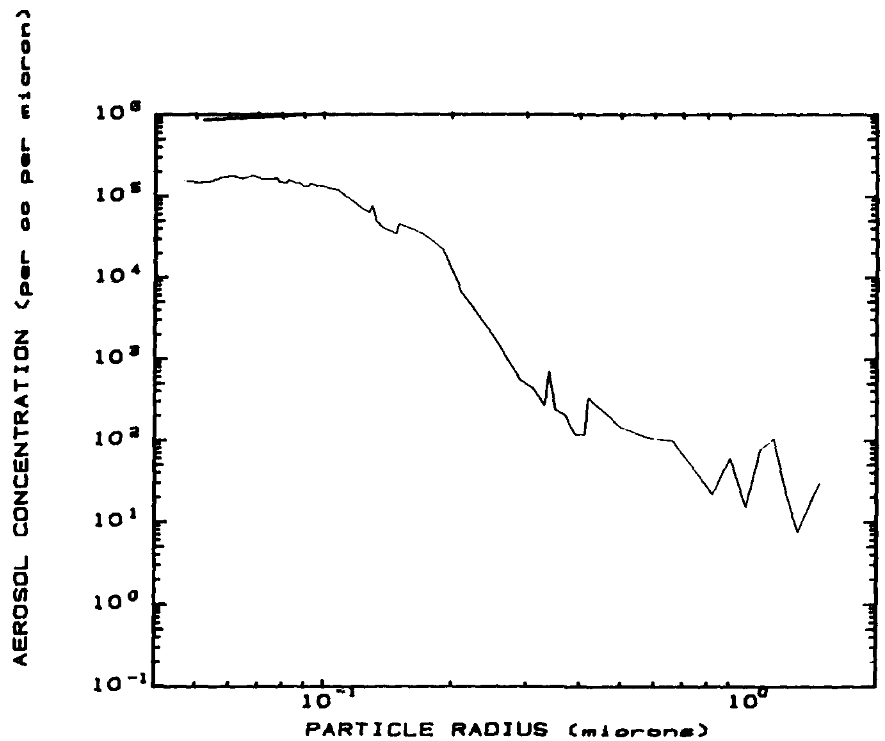


Fig. 1. Aerosol particle concentration per cc per micron as a function of particle radius, (corrected). Mace Head, 11 Dec. 1987. Time: 0330-0911. Wind direction: easterly.

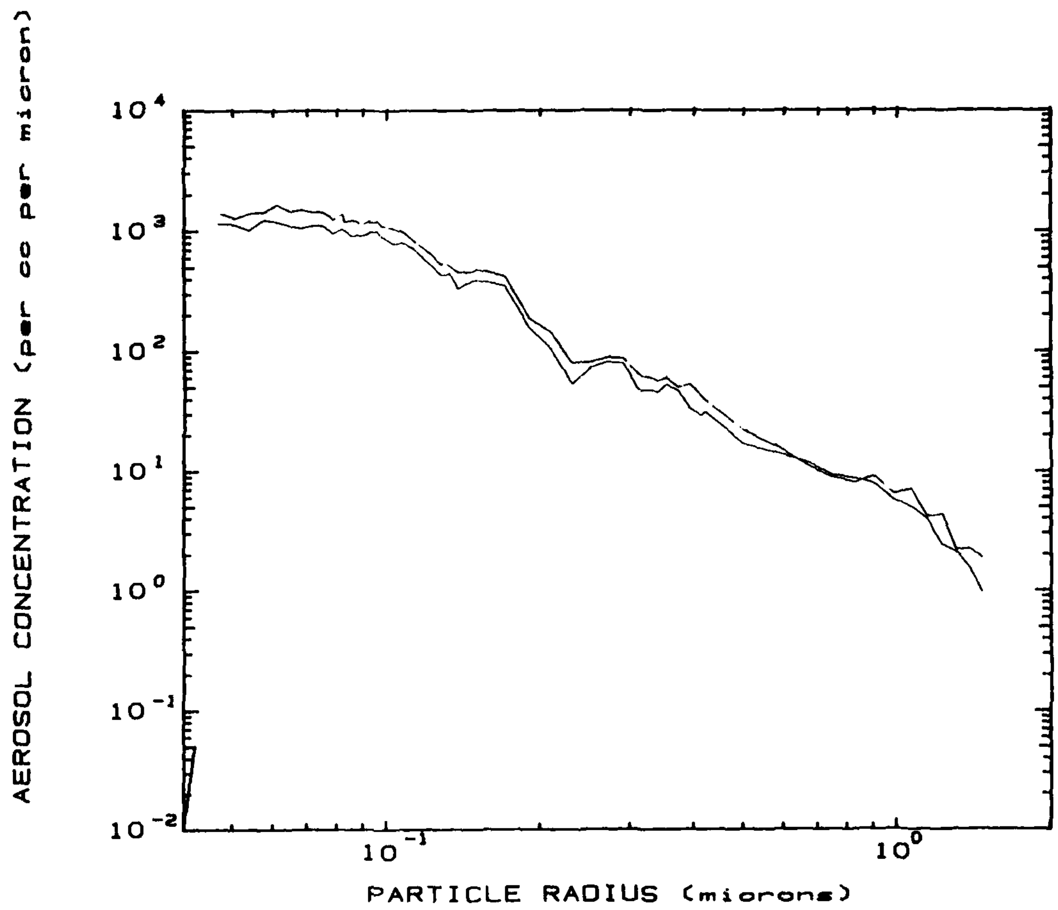


Fig. 2. Aerosol particle concentration per cc per micron as a function of particle radius. Mace Head, 17 Dec. 1987. Time: 1511-1516 (upper curve); 1431-1436 (lower curve). Wind direction: southerly.

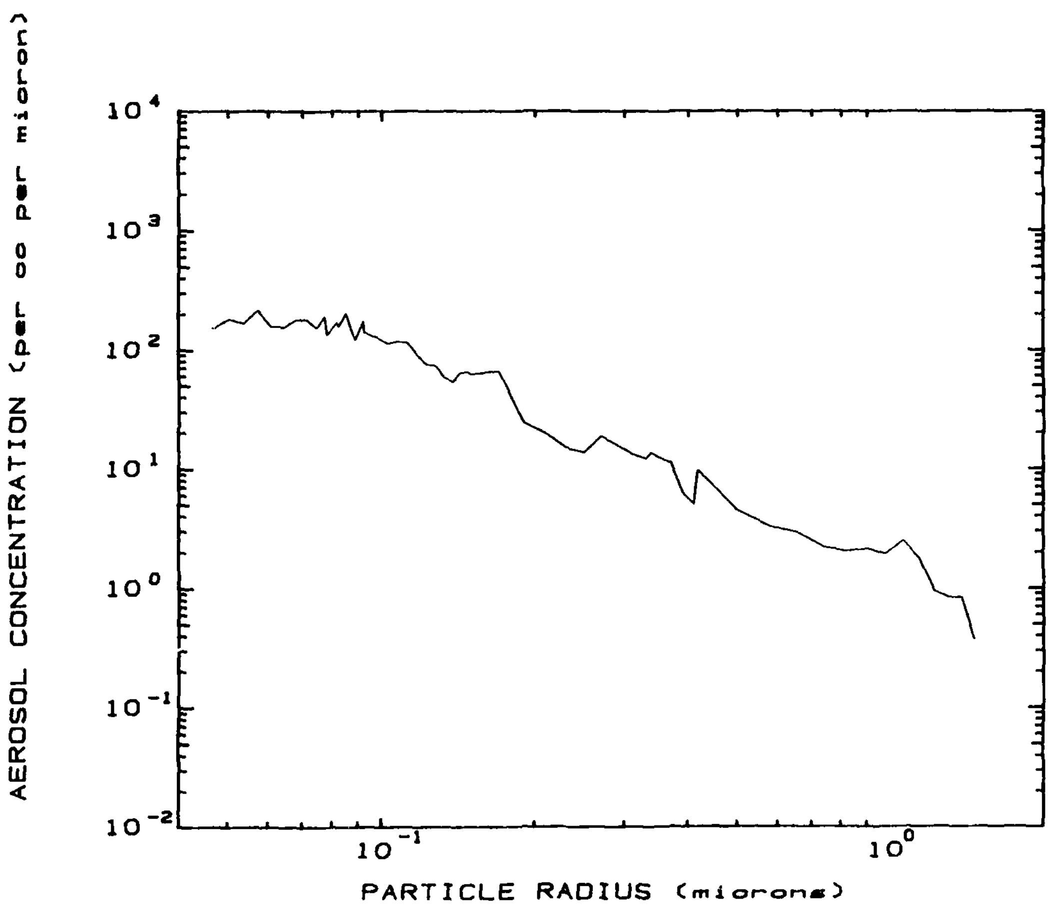


Fig. 3. Aerosol particle concentration per cc per micron as a function of particle size. Mace Head 14 January 1988. Time 1140-1145. Wind direction: south westerly.

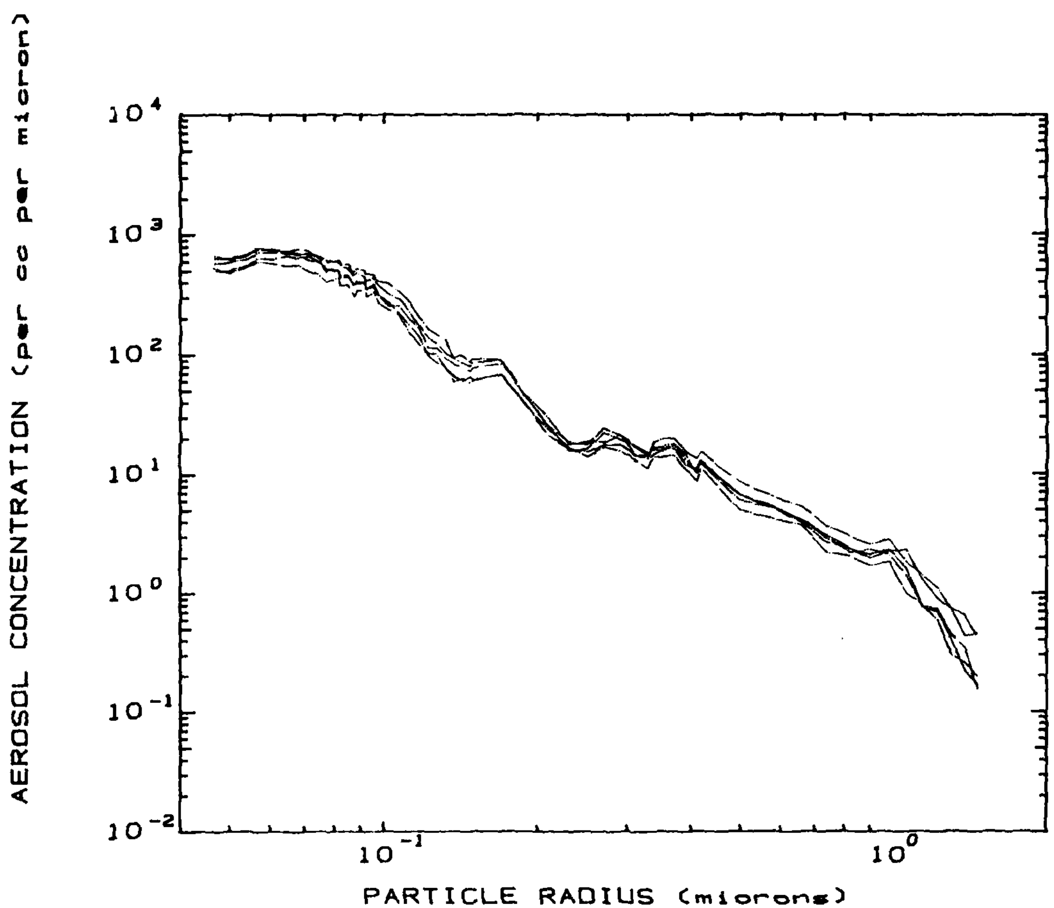


Fig. 4. *Five consecutive data cycles of particle number concentration per cc per micron as a function of particle radius.*
Mace Head, 10-11 March 1988. Time: 2230-0235. Wind direction: west south westerly.

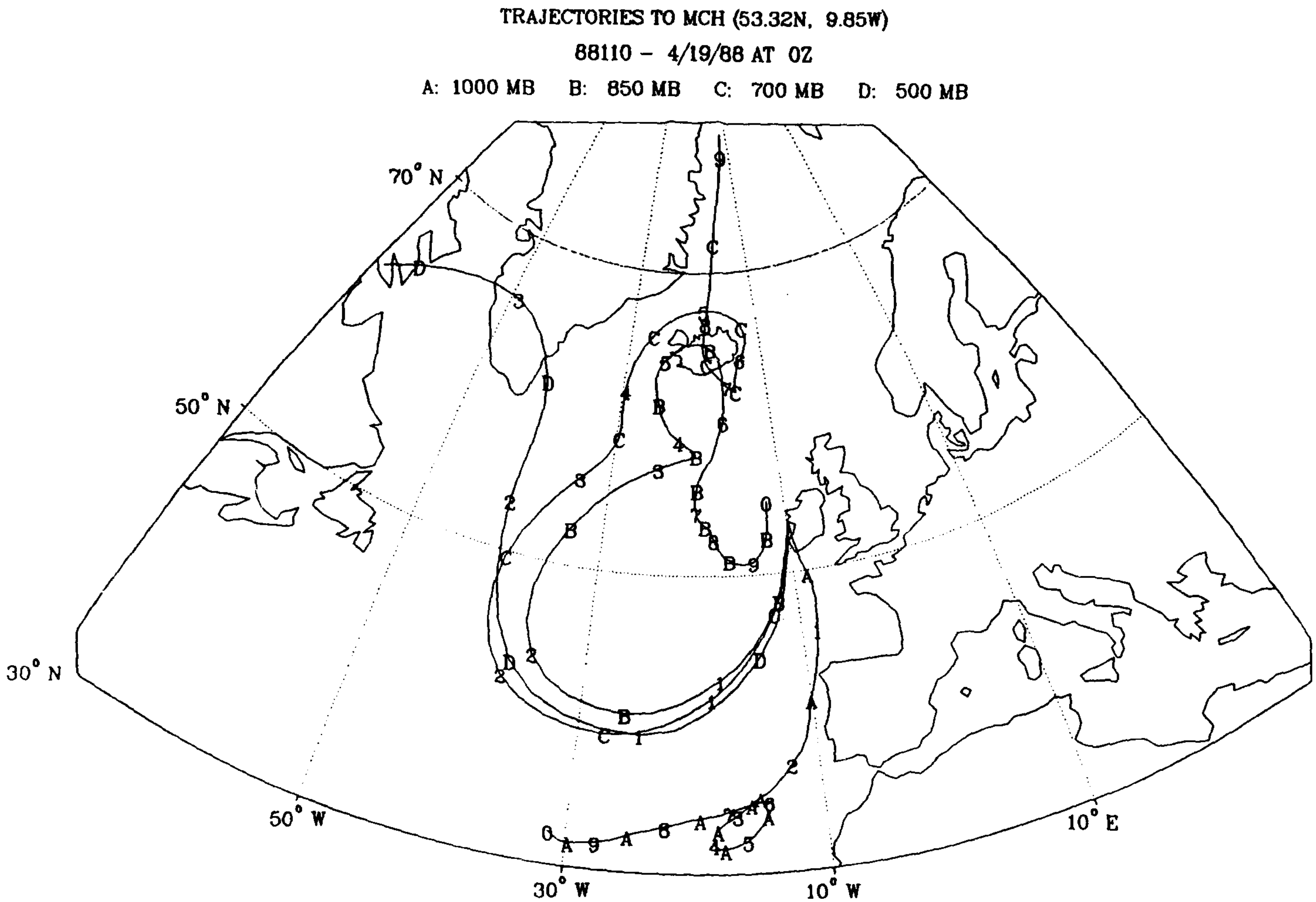


Fig. 5 Atmospheric air trajectories to Mace Head (MCH) on 19 April 1988 at Time 0Z, for pressure levels of 1000 mb, 850 mb, 700 mb and 500 mb.

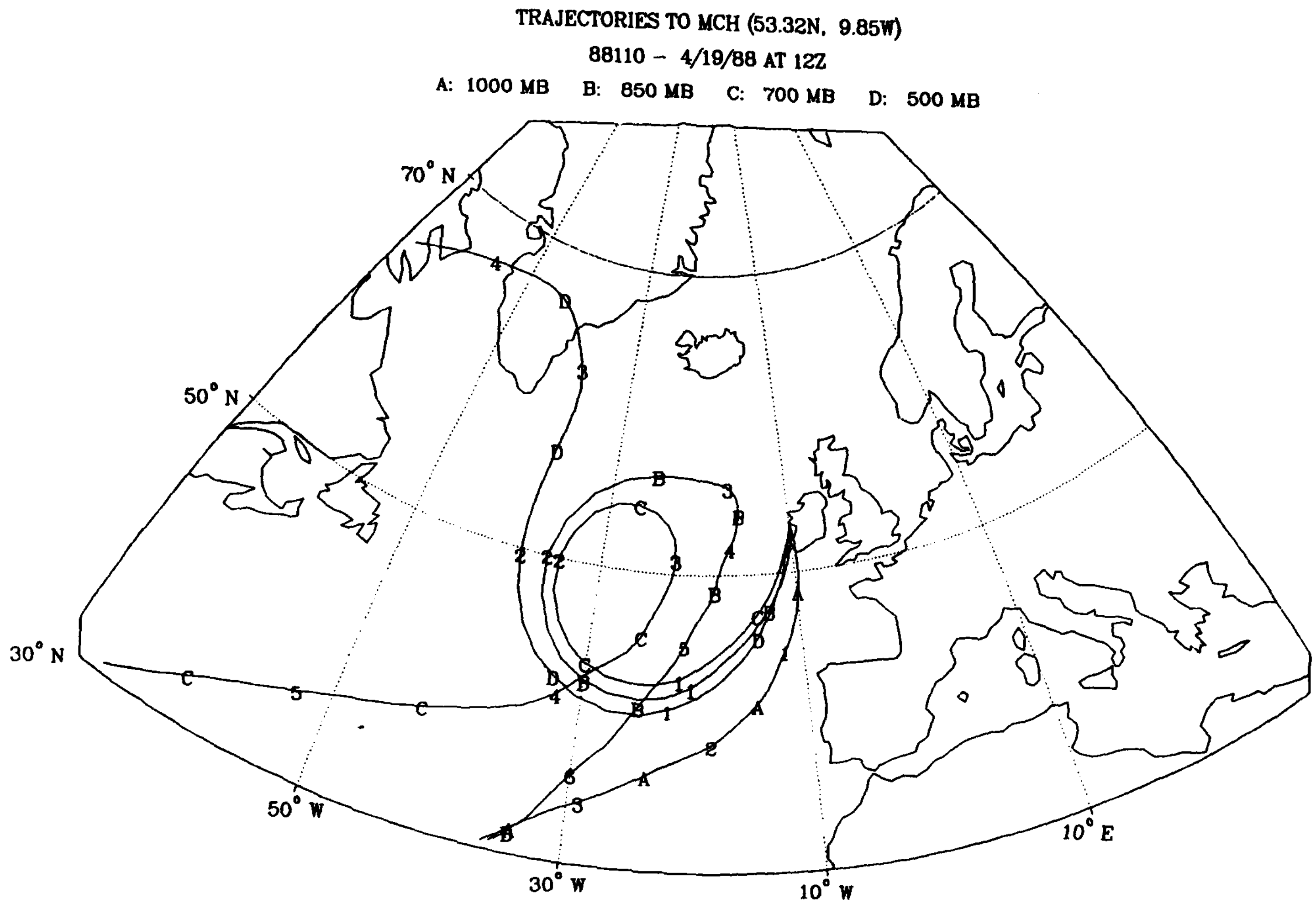


Fig. 6 As per Fig. 5 except for 12Z.

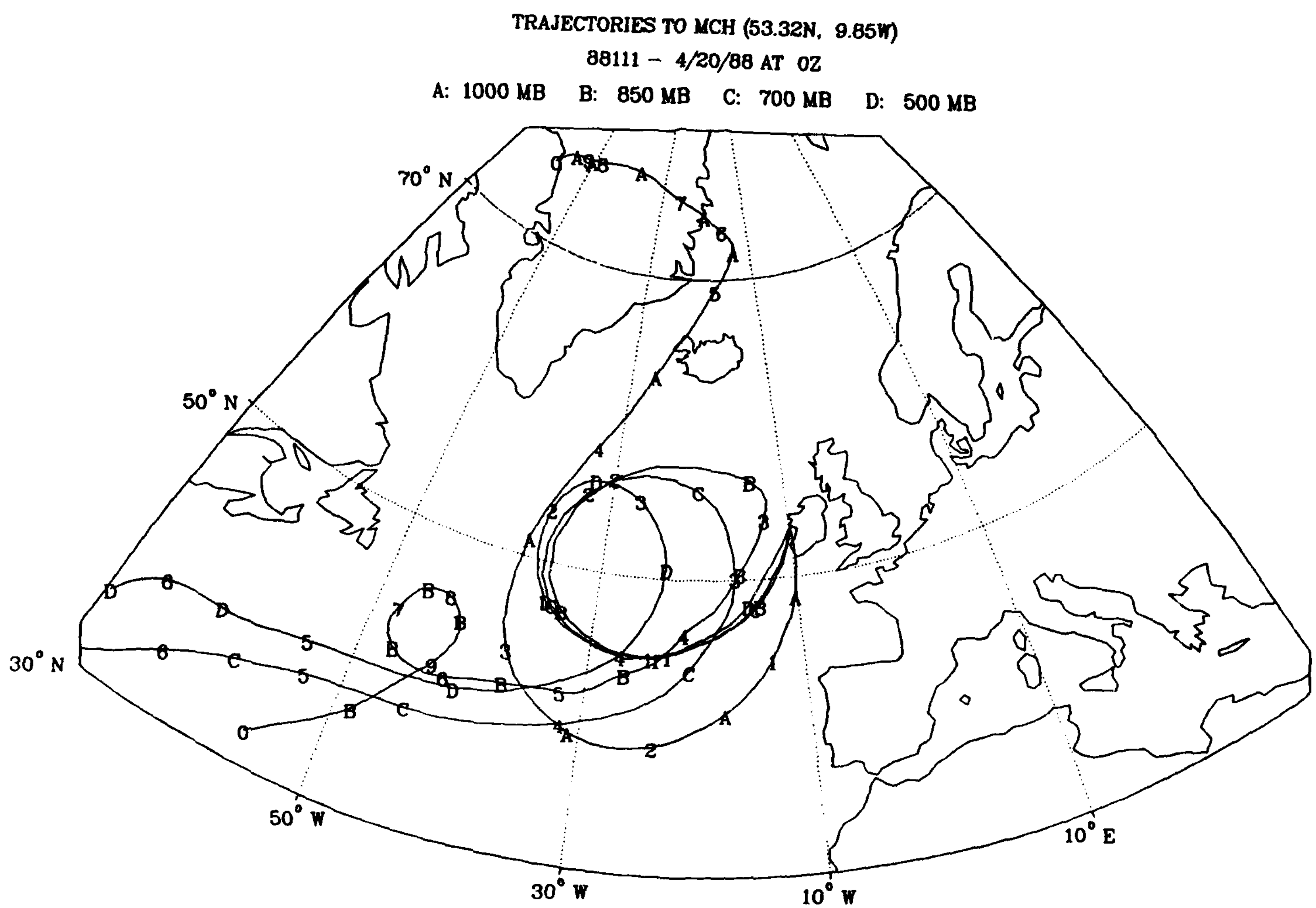


Fig. 7 As per Fig. 5 except for 20 April 1988

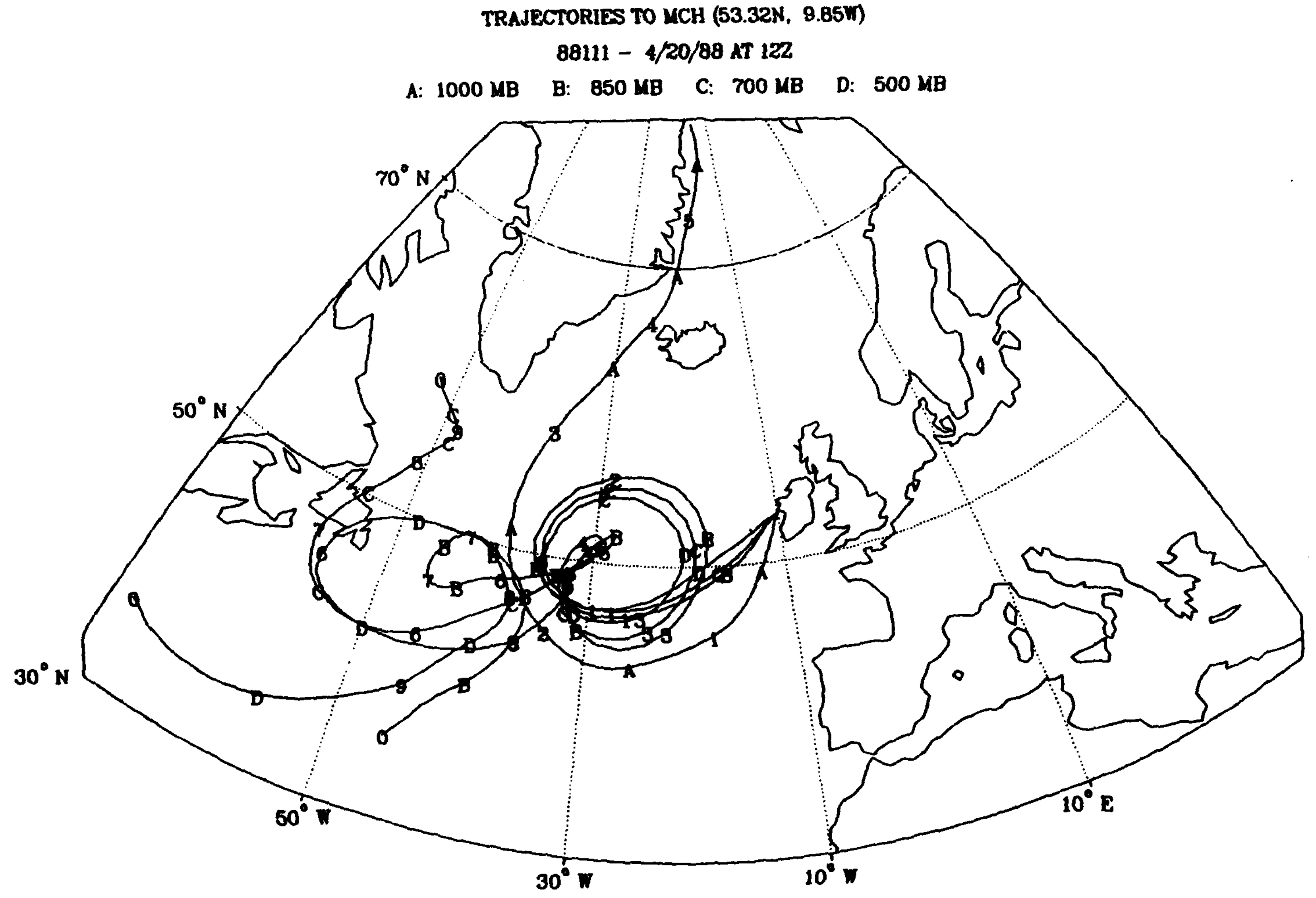


Fig. 8 As per Fig. 6 except for 20 April 1988

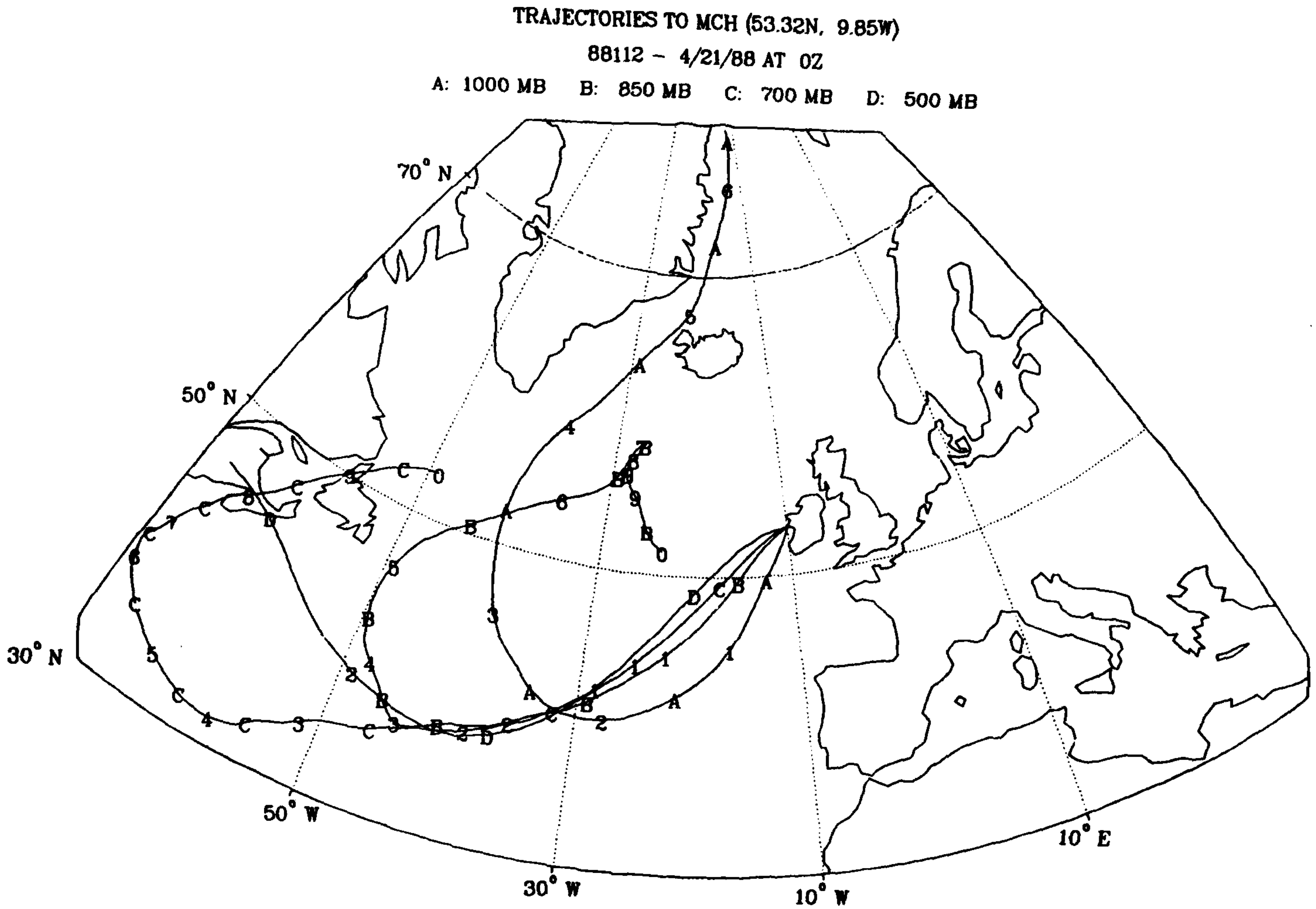


Fig. 9. As per Fig. 5 except for 21 April 1988

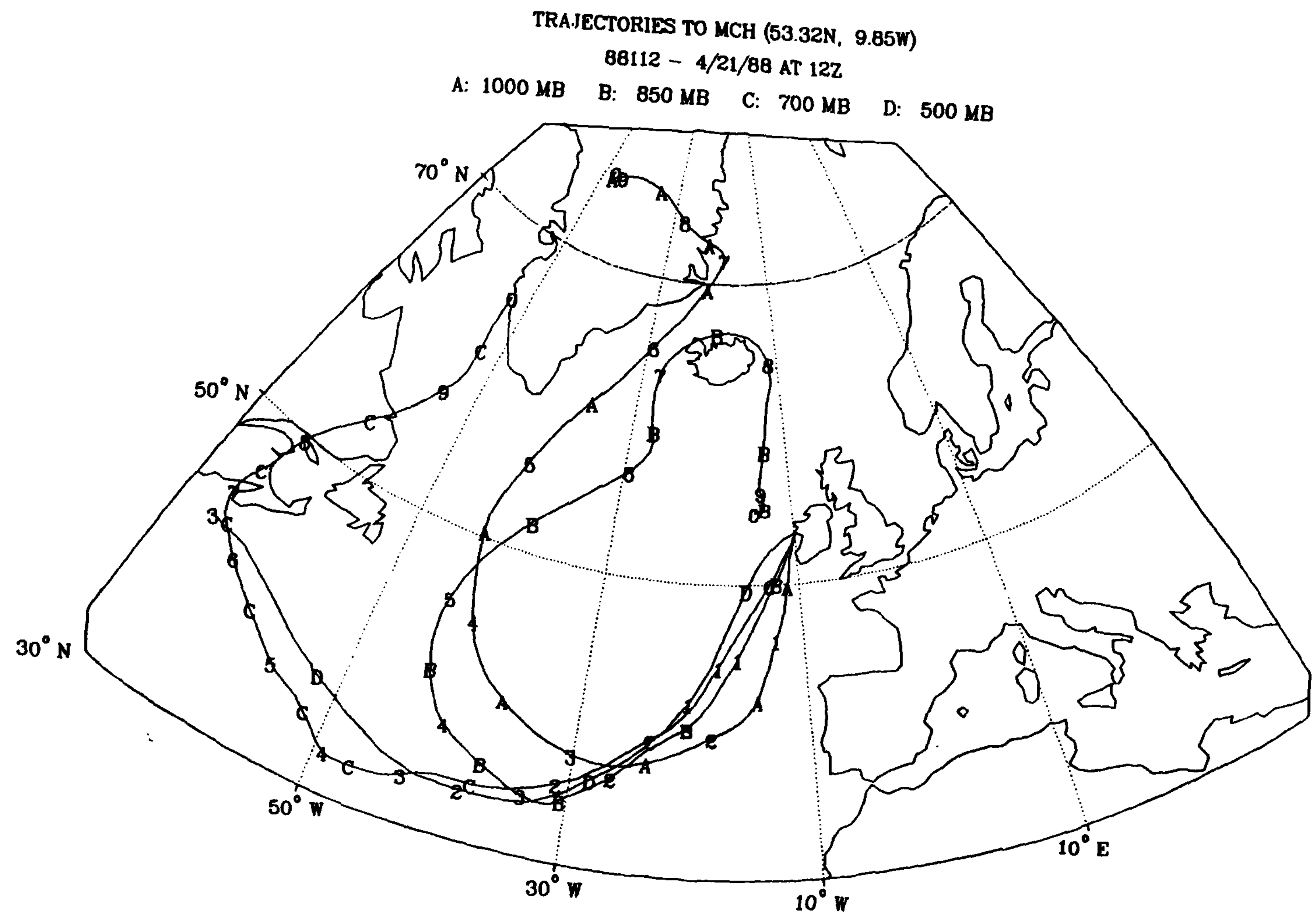


Fig. 10 As per Fig. 6 except for 21 April 1988

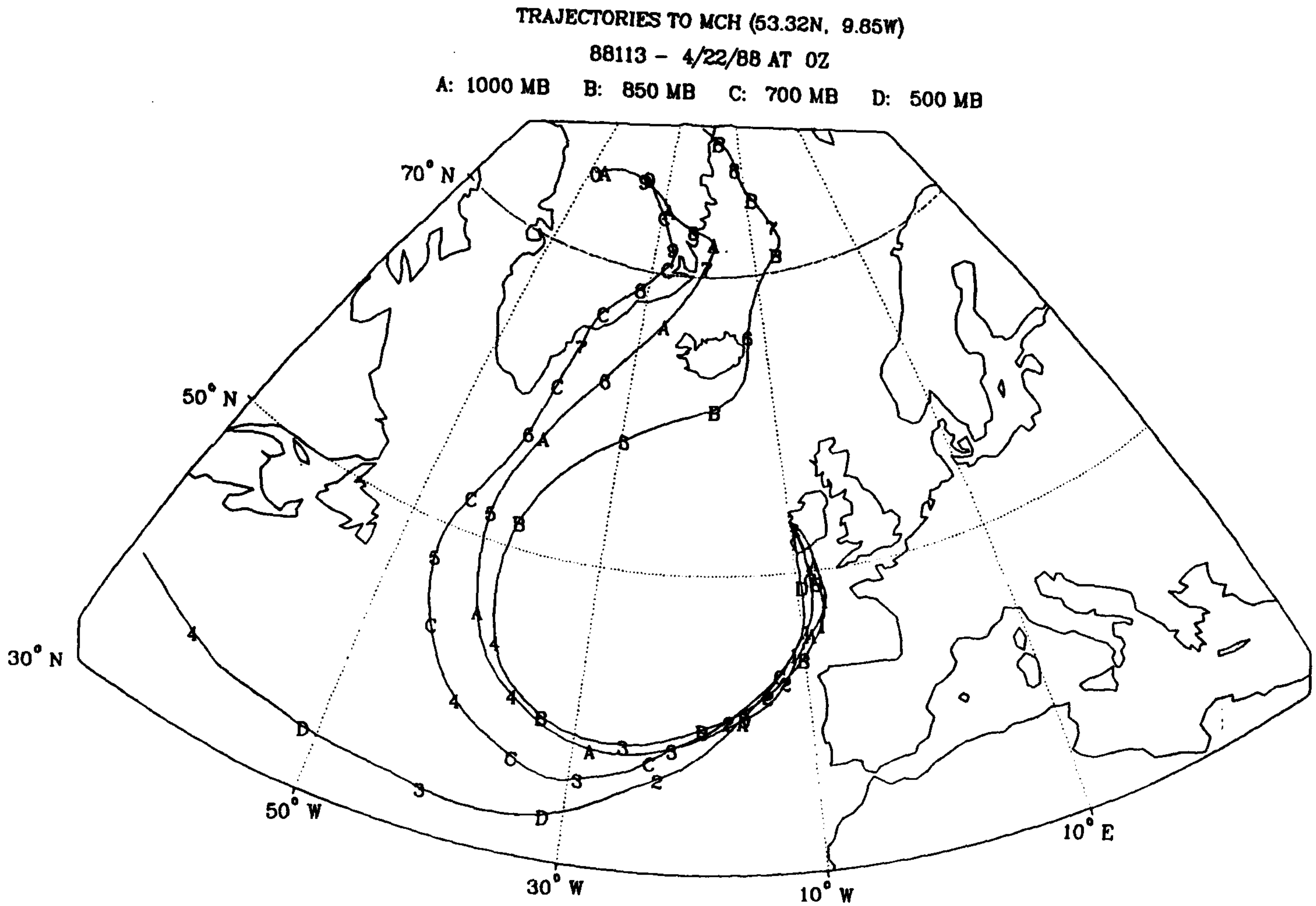


Fig. 11 As per Fig. 5 except for 22 April 1988

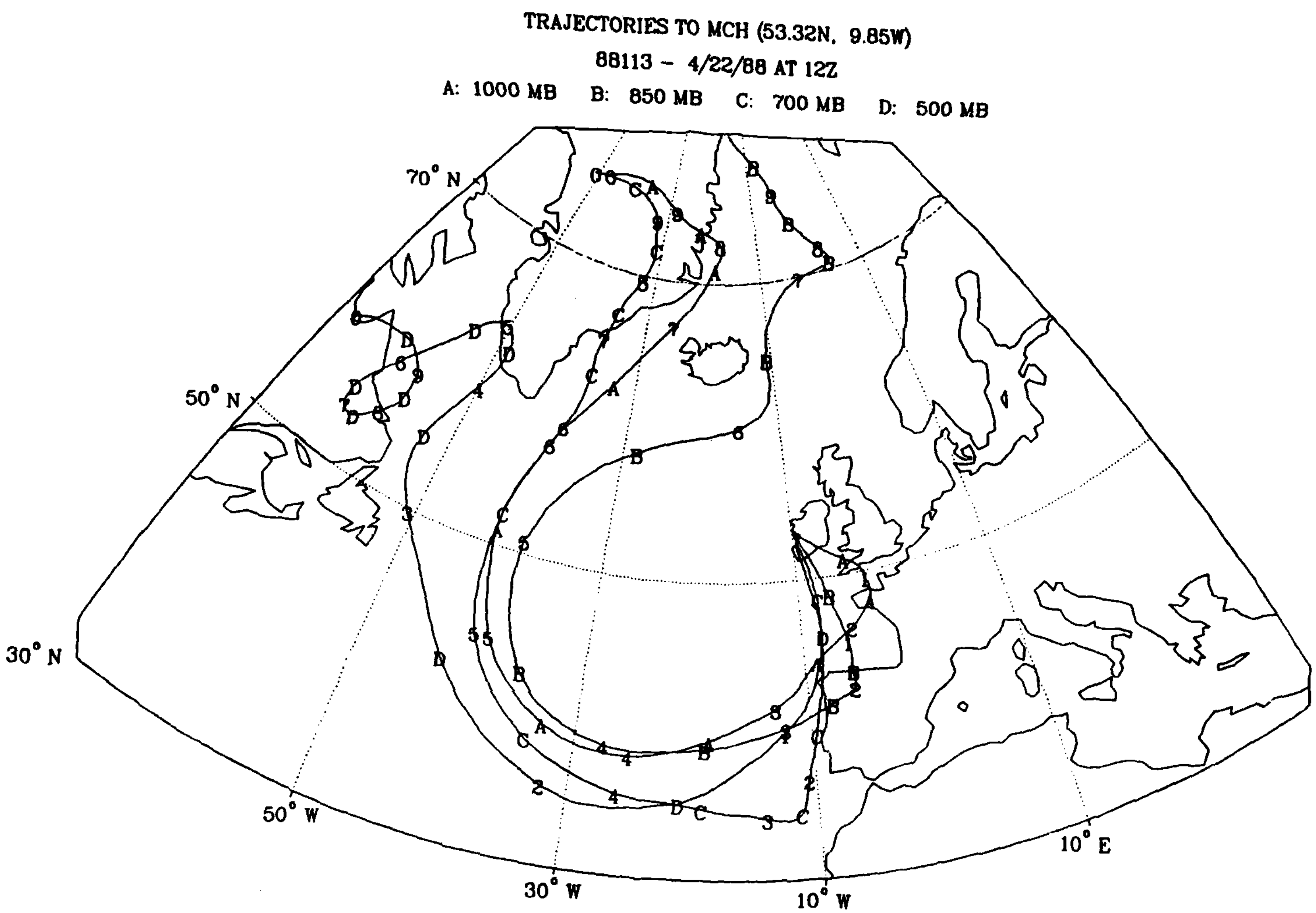


Fig. 12 As per Fig. 6 except for 22 April 1988

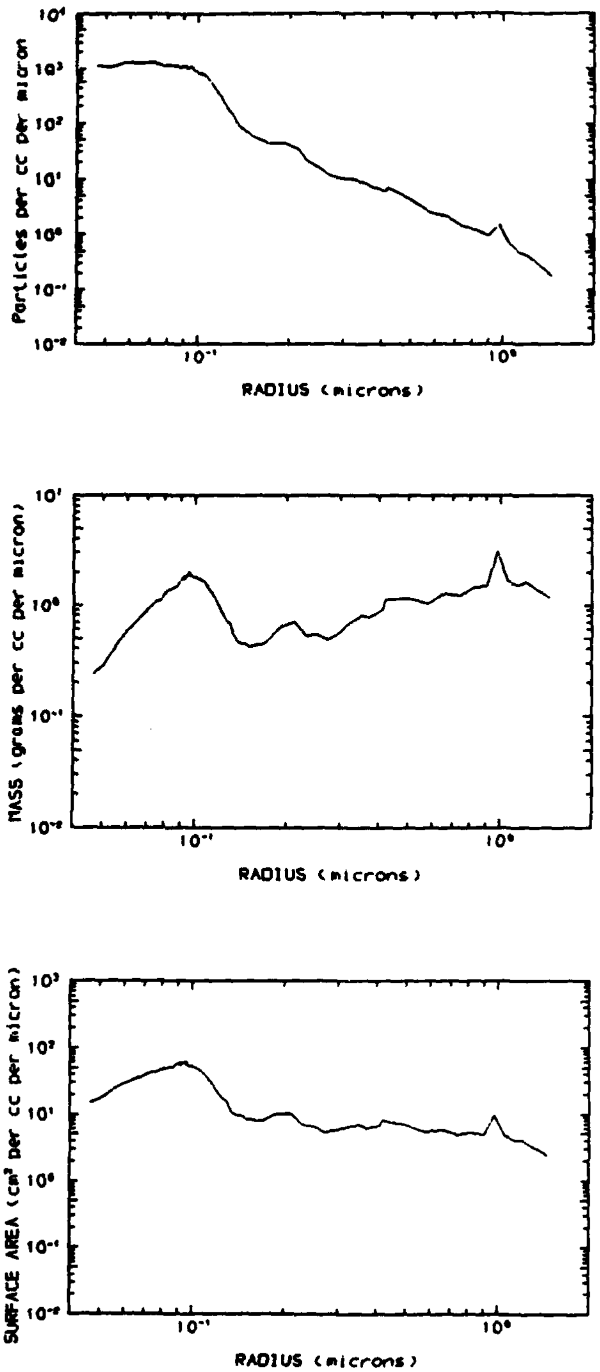


Fig. 13 Particle number concentration per cm^3 per micron (upper plot), particle mass concentration in grams per cm^3 per micron (centre plot) and particle surface area in cm^2 per cm^3 per micron (lower plot) as a function of particle radius in microns. Measurement period : April 19, 1988; 1330 - 1600 local time.

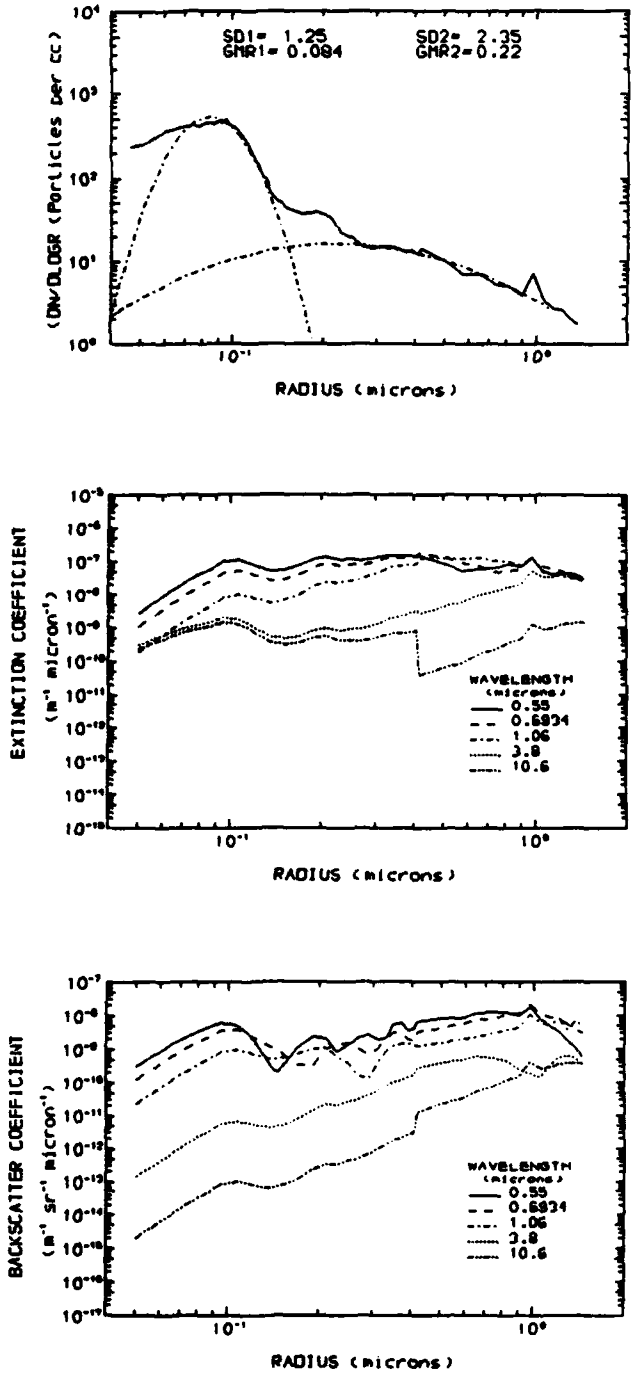


Fig. 14 Particle size distribution (upper curve) plotted in the form dN/dlogr versus particle radius r for the data of April 19, 1988; 1330 - 1600 local time. Two modes with log-normal parameter values for geometric mean radius (GMR) r_g and geometric standard deviation (SD) σ_g are fitted to the data.

Calculated extinction (centre plot) and backscatter (lower plot) coefficients, based on the measured data shown in the previous figure are plotted as a function of particle radius for radiation wavelengths 0.55, 0.6934, 1.06, 3.8 and 10.6 μm. It was assumed that particles with radius < 0.4 μm consisted of Ammonium Sulphate and particles with radius > 0.4 μm consisted of Sodium Chloride.

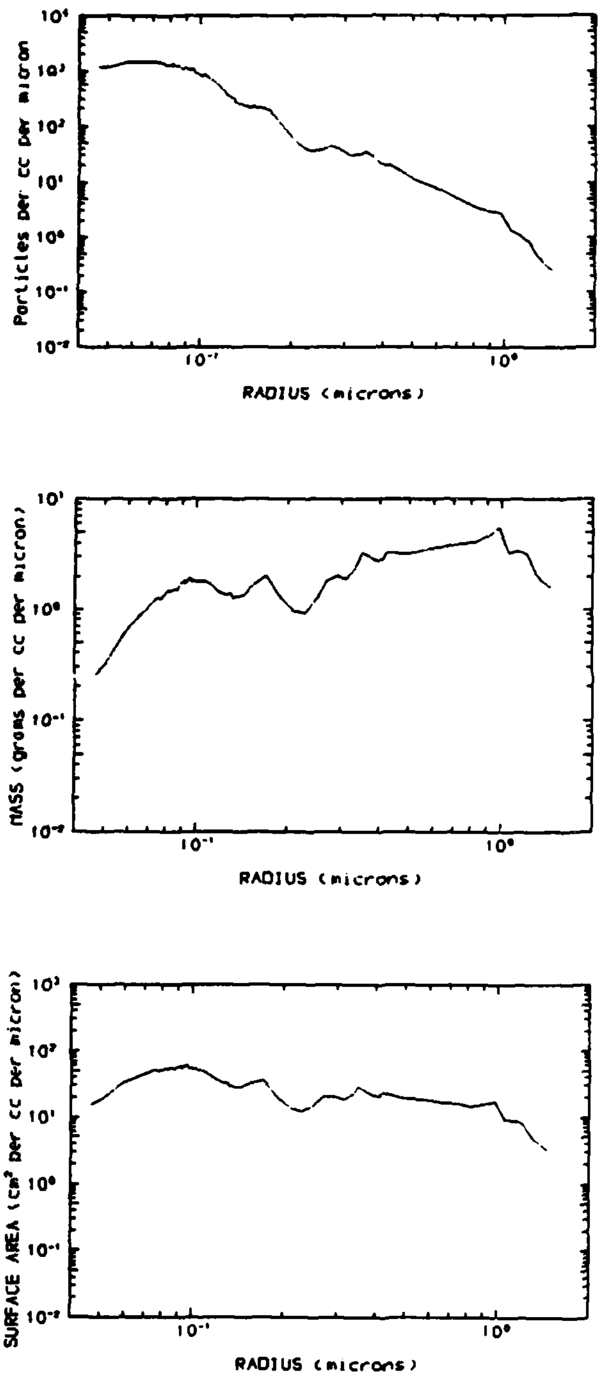


Fig. 15 As per Fig. 13 except for the measurement period: April 20, 1988, 1800 - 2400 local time.

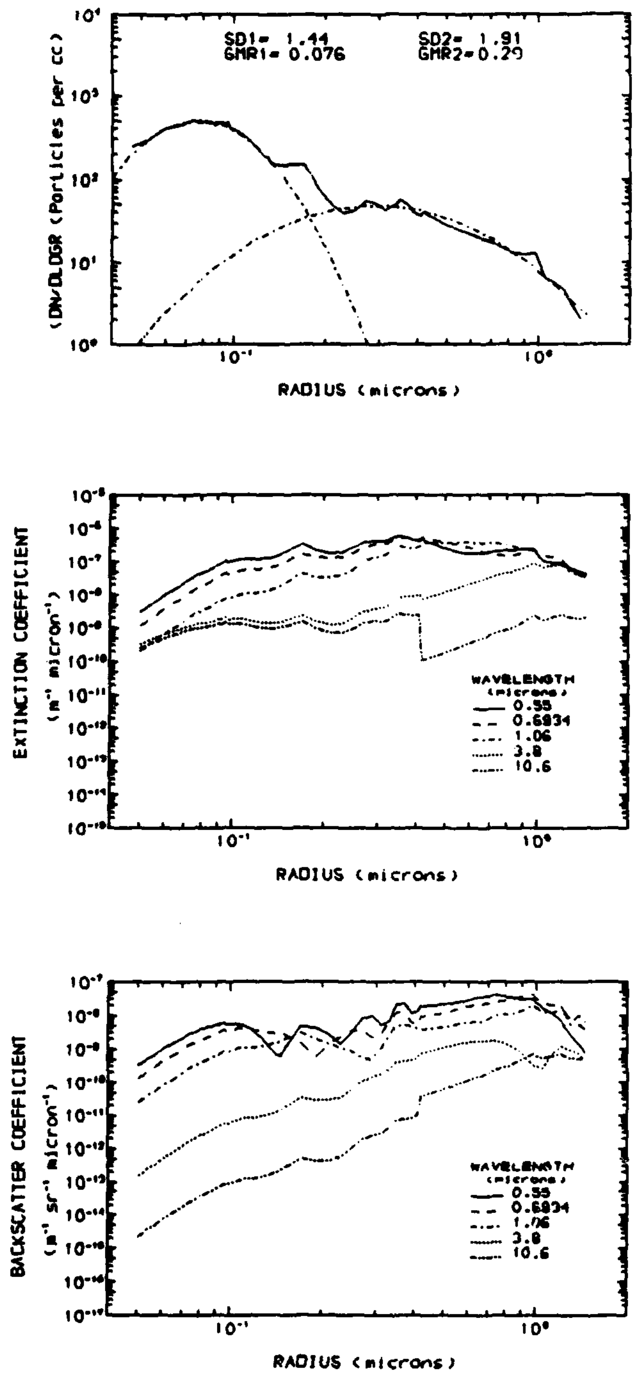


Fig. 16 As per Fig. 14 except for the measurement period : April 20, 1988; 1800 - 2400 local time.

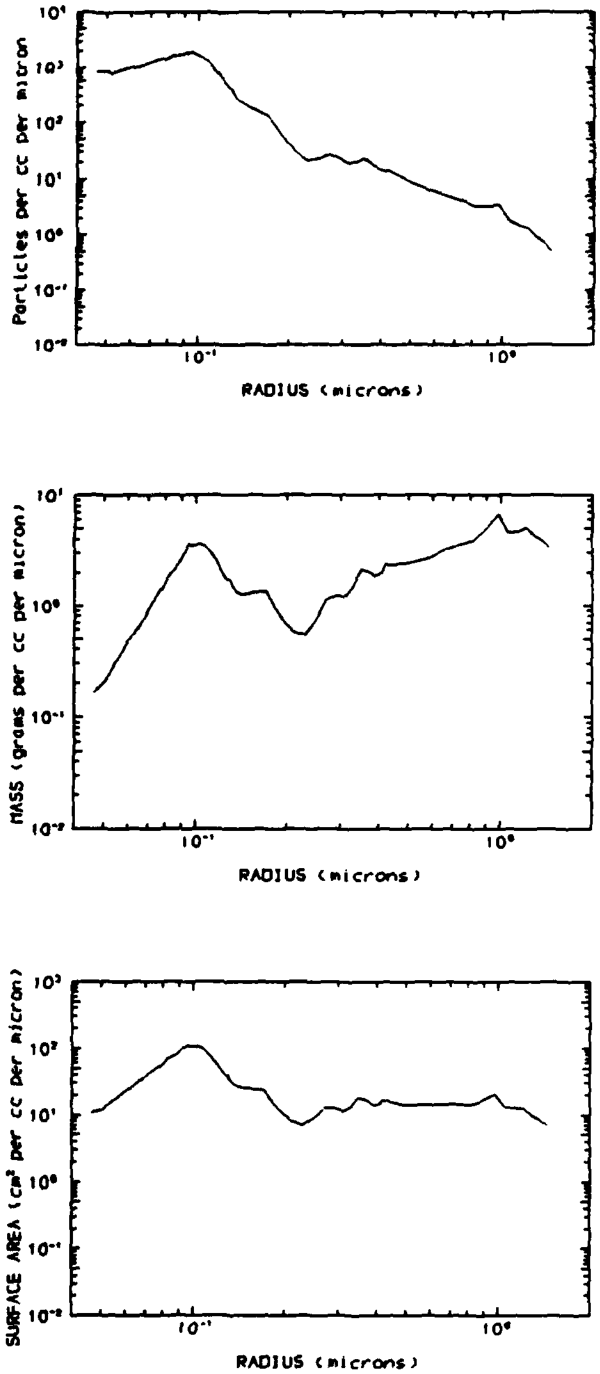


Fig. 17 As per Fig. 13 except for the measurement period : April 21, 1988; 0150 - 1245 local time.

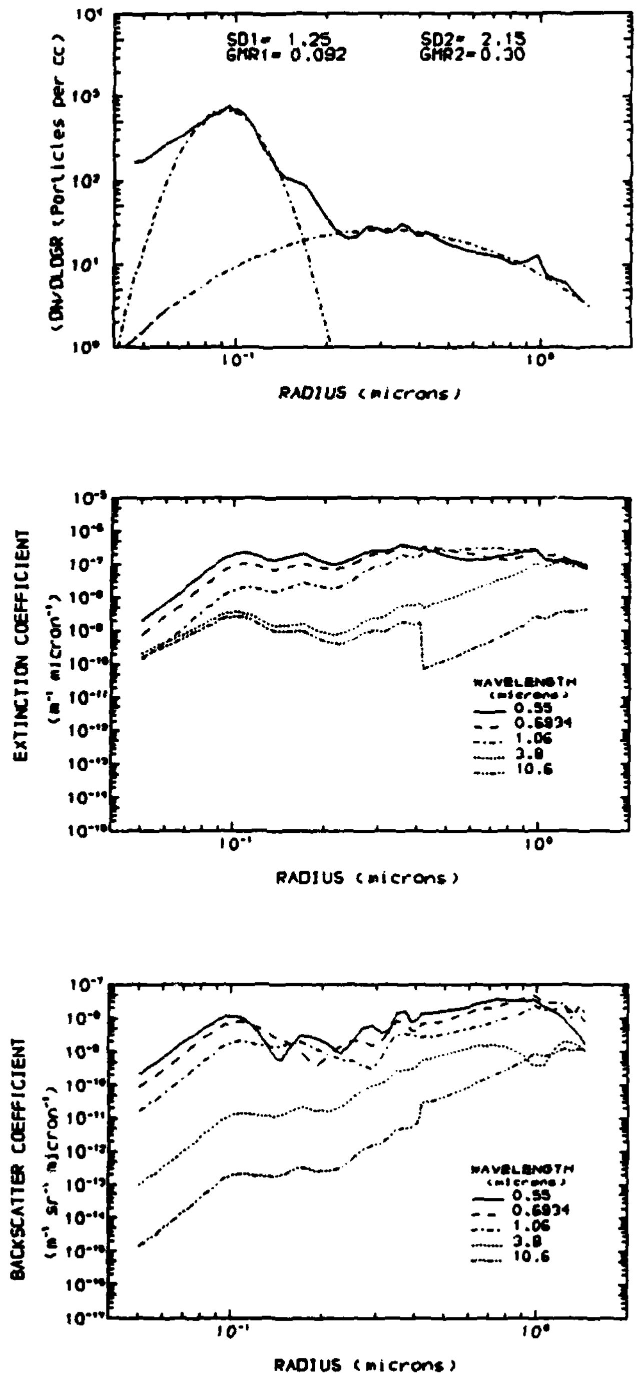


Fig. 18 As per Fig. 14 except for the measurement period : April 21, 1988; 0150 - 1245 local time.

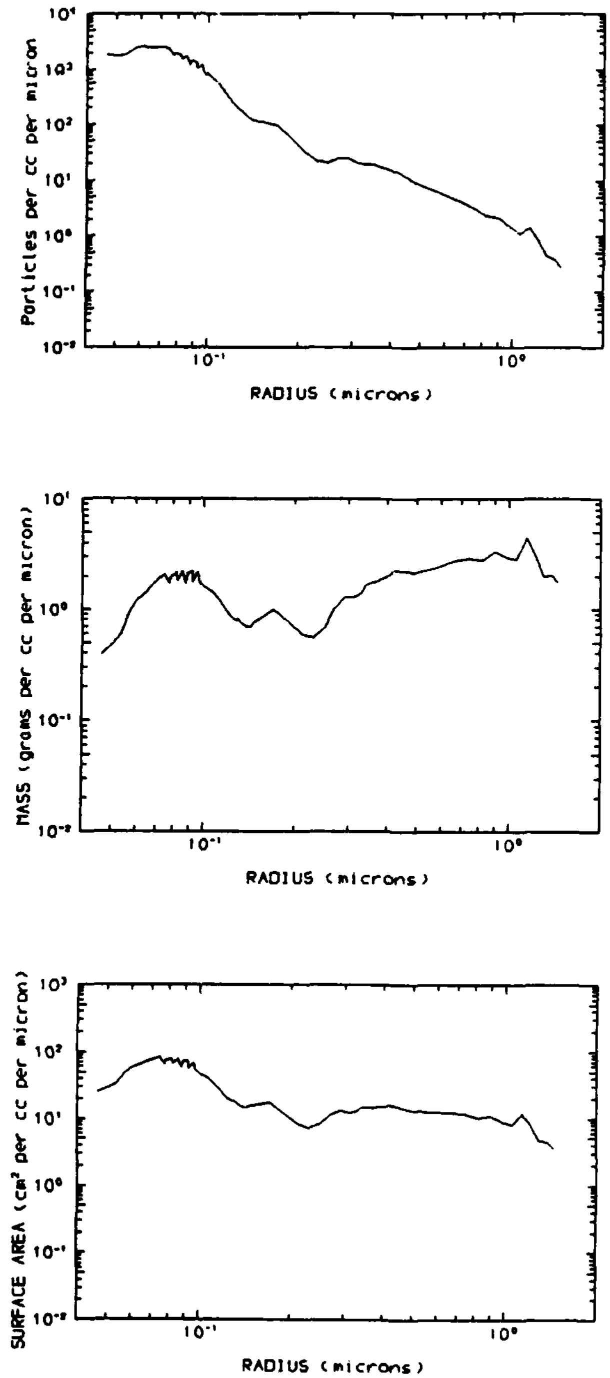


Fig. 19 As per Fig. 13 except for the measurement period : June 1, 1988; 1850 -2150 local time.

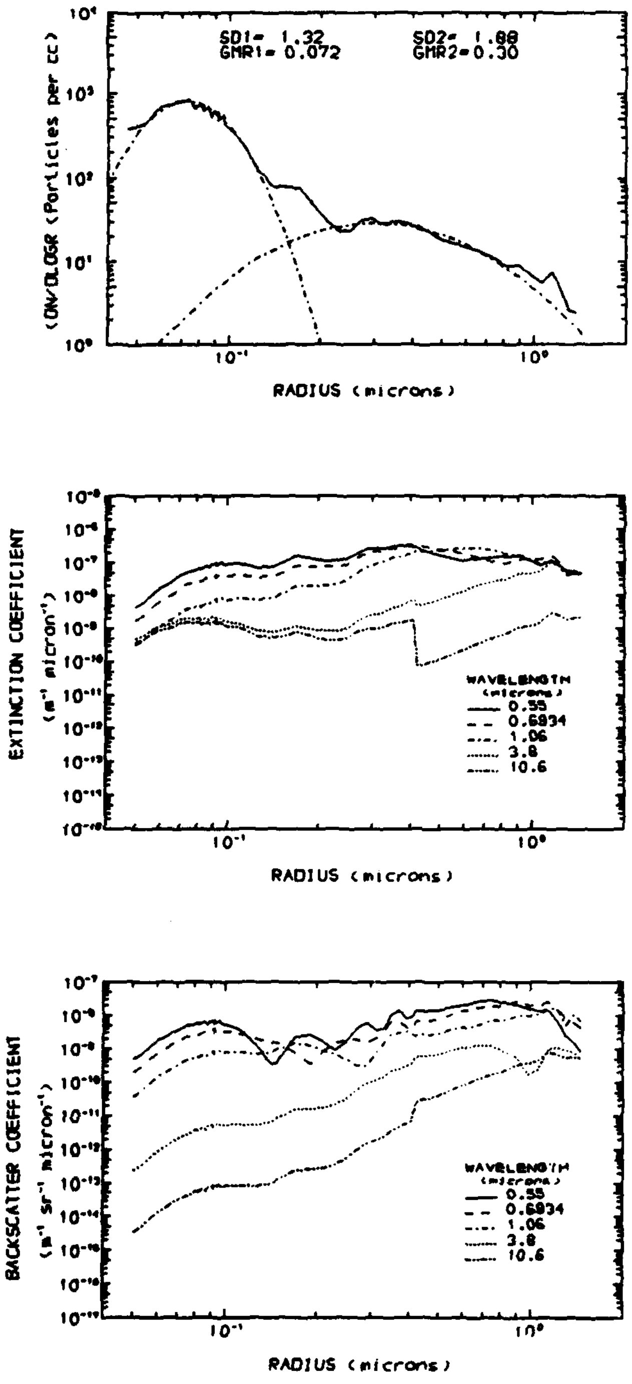


Fig. 20 As per Fig. 14 except for the measurement period : June 1, 1988; 1850 - 2150 local time.

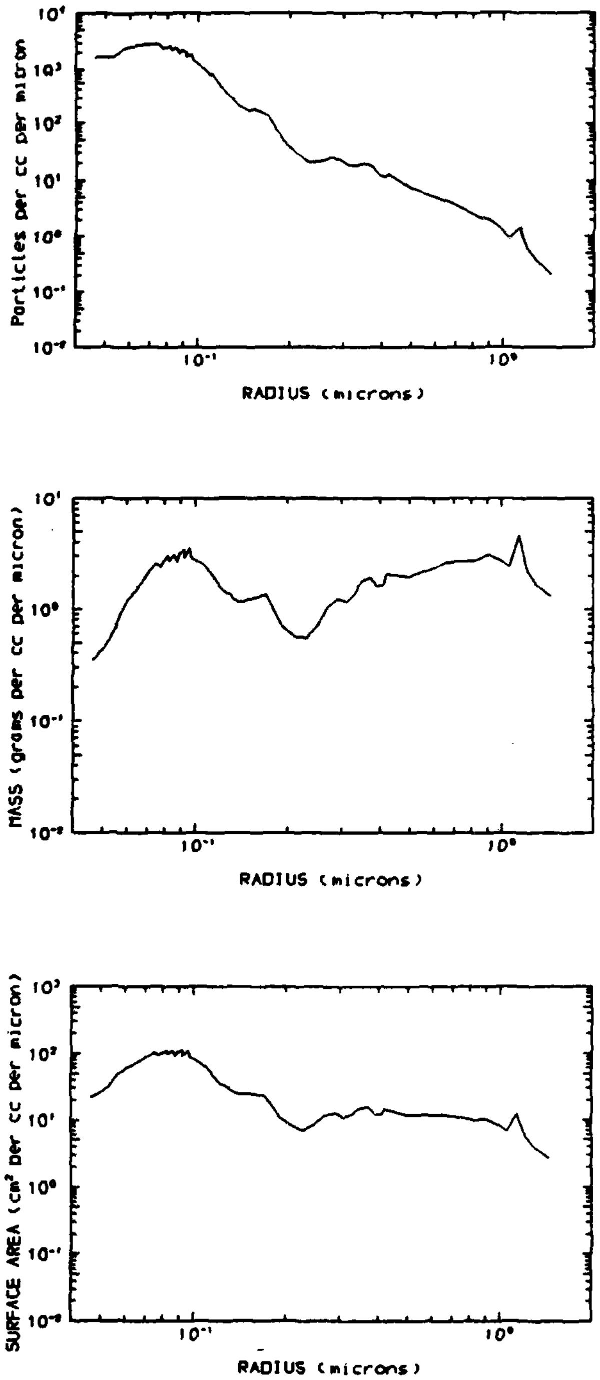


Fig. 21 As per Fig. 13 except for the measurement period : June 2, 1988; 0050 -1000 local time.

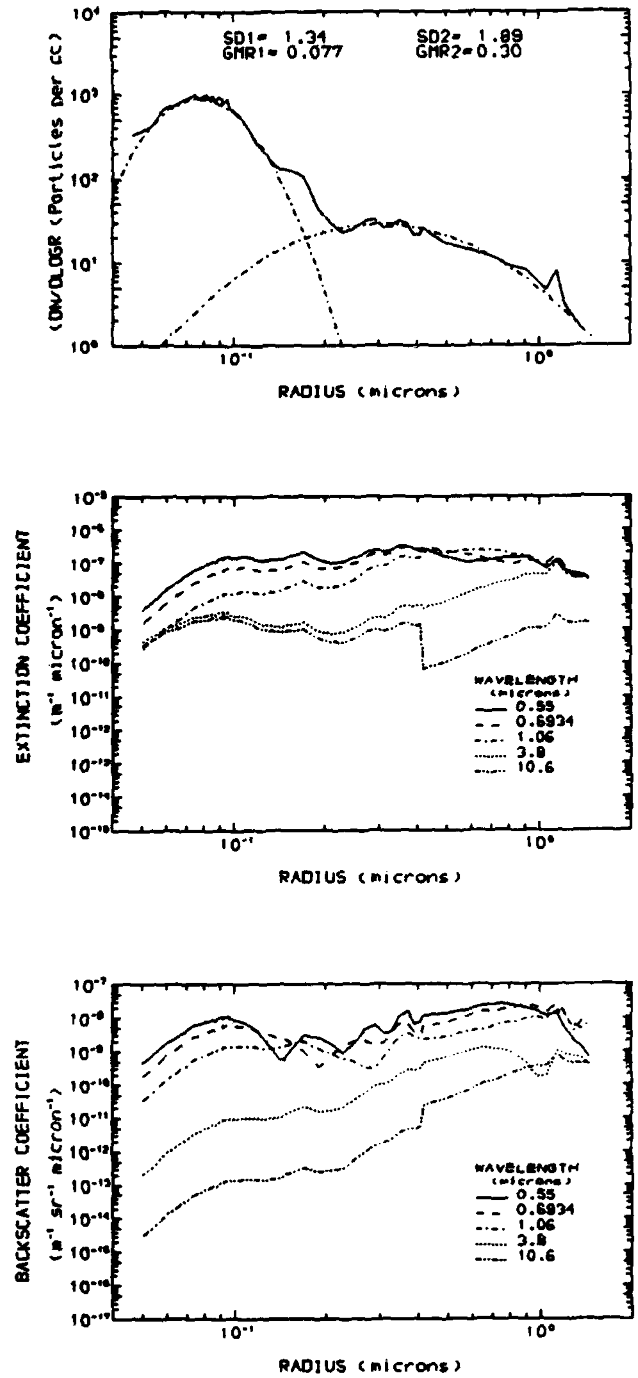


Fig. 22 As per Fig. 14 except for the measurement period : June 2, 1988; 0050 - 1000 local time.

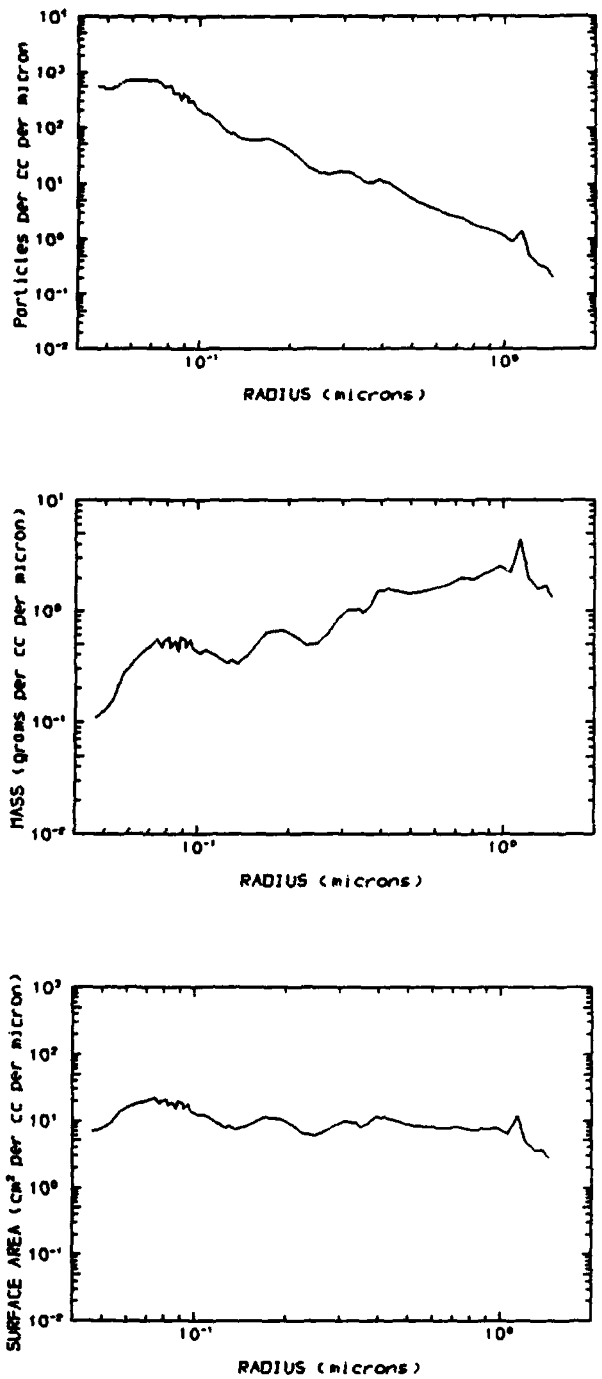


Fig. 23 As per Fig. 13 except for the measurement period : June 2, 1988; 1600 - 2000 local time.

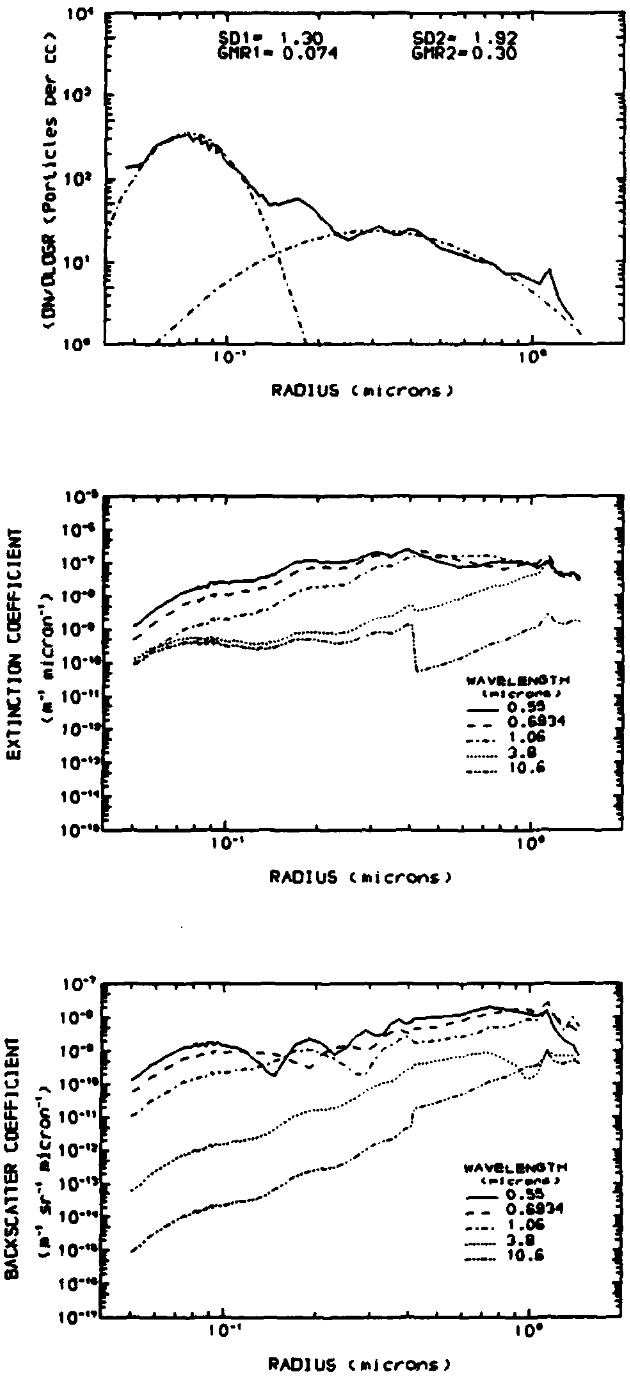


Fig. 24 As per Fig. 14 except for the measurement period : June 2, 1988; 1600 - 2000 local time.

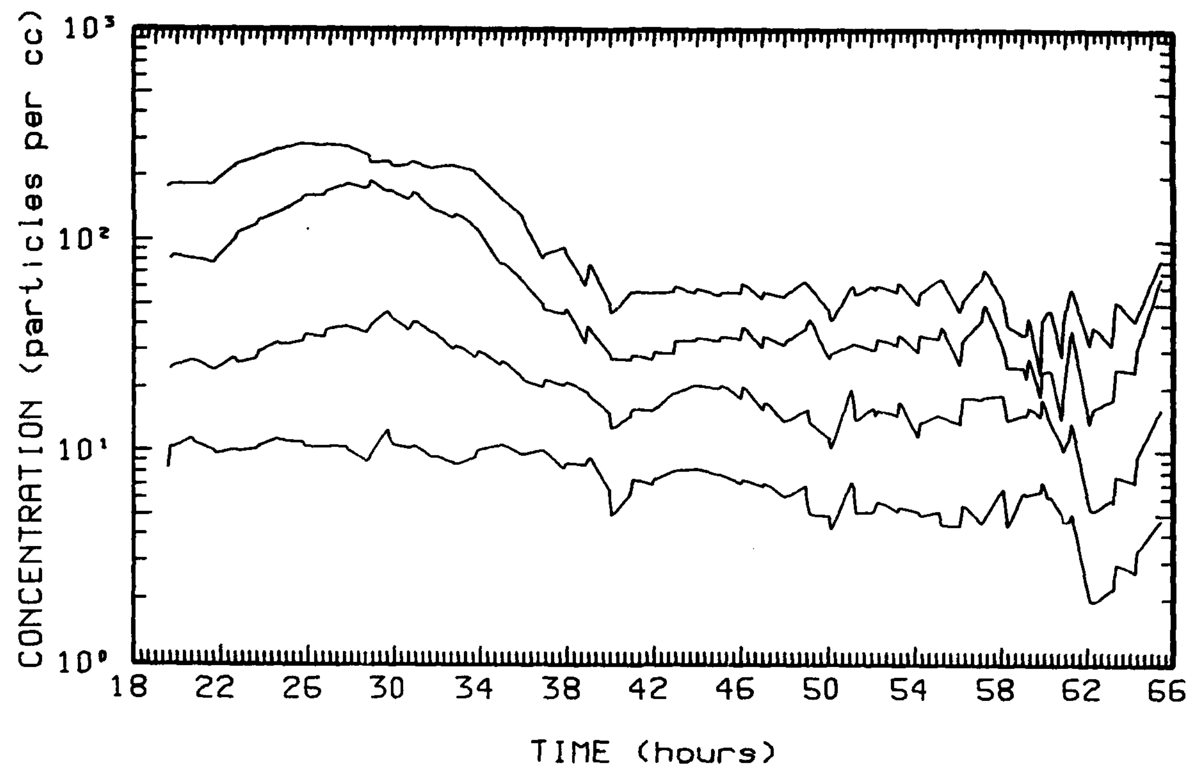


Fig. 25 Durnal variation in aerosol particle concentration for period from 1850 June 1 to 1550 June 3, 1988. Ranges 3 through 0 are shown from the top curve to the lowest curve.

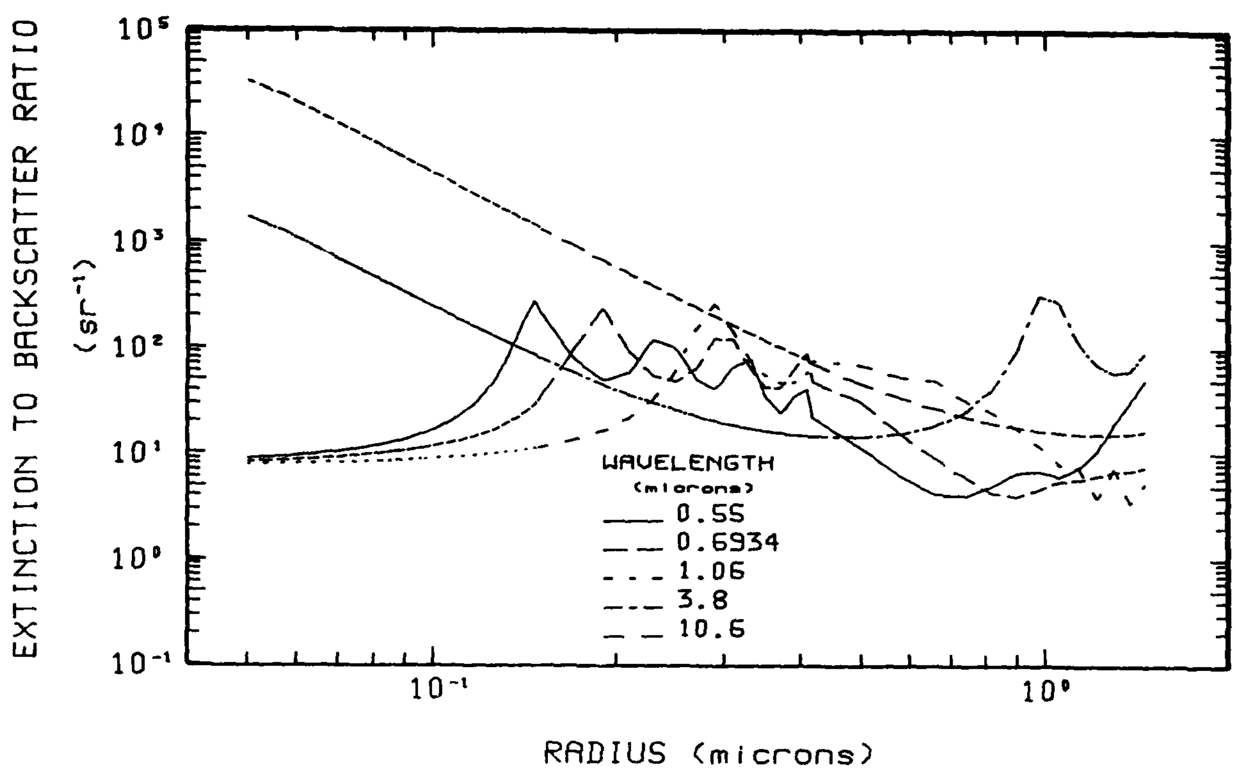


Fig. 26 Extinction to Backscatter ratio per sr as a function of particle radius and wavelength. Constituent : Ammonium Sulphate.

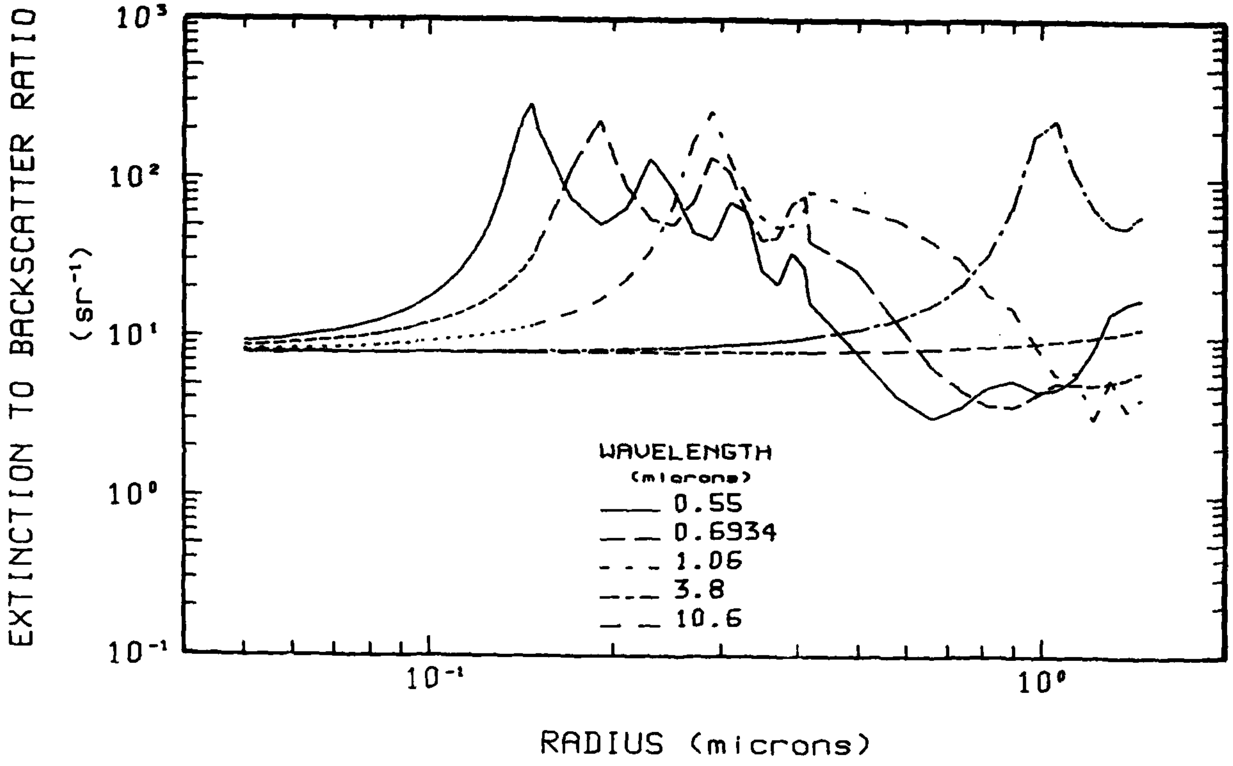


Fig. 27 As per Fig. 26 except for constituent : Sodium Chloride

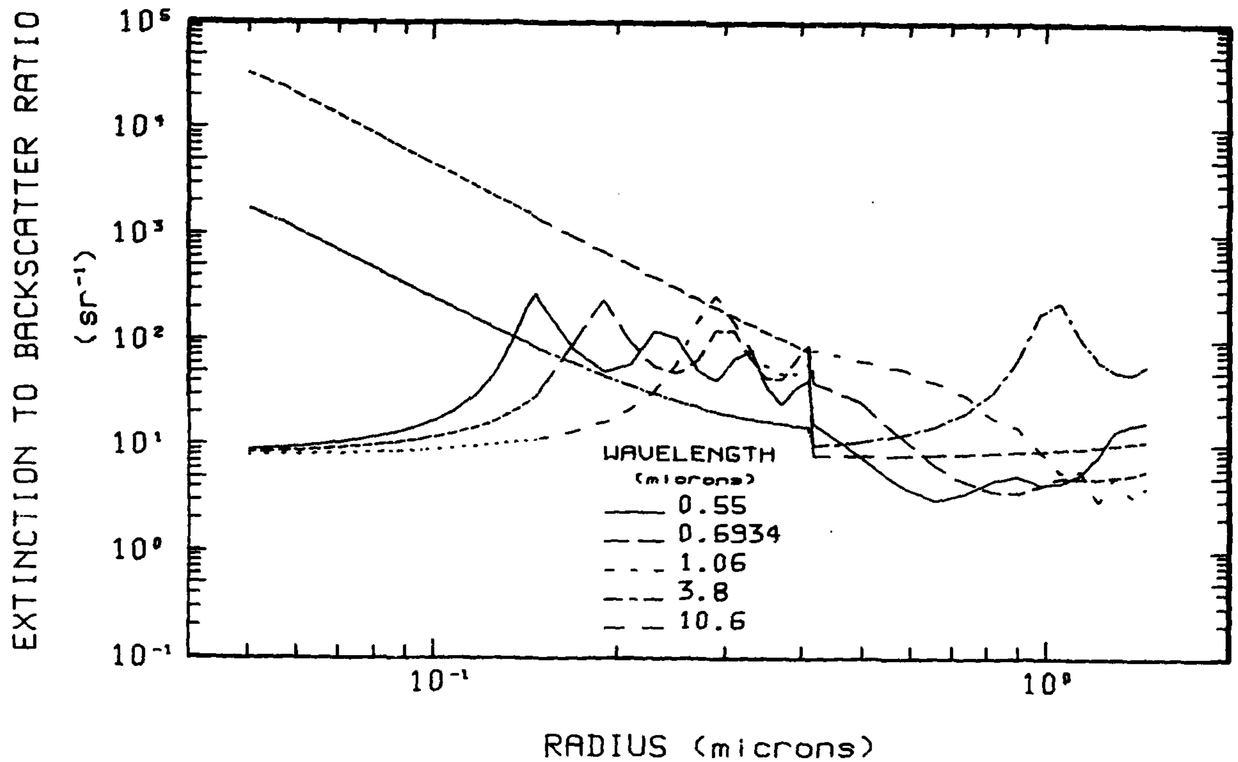


Fig. 28 As per Fig. 26 except for constituents : Ammonium Sulphate (for radius < 0.4 μm) and sodium chloride (for radius > 0.4 μm).

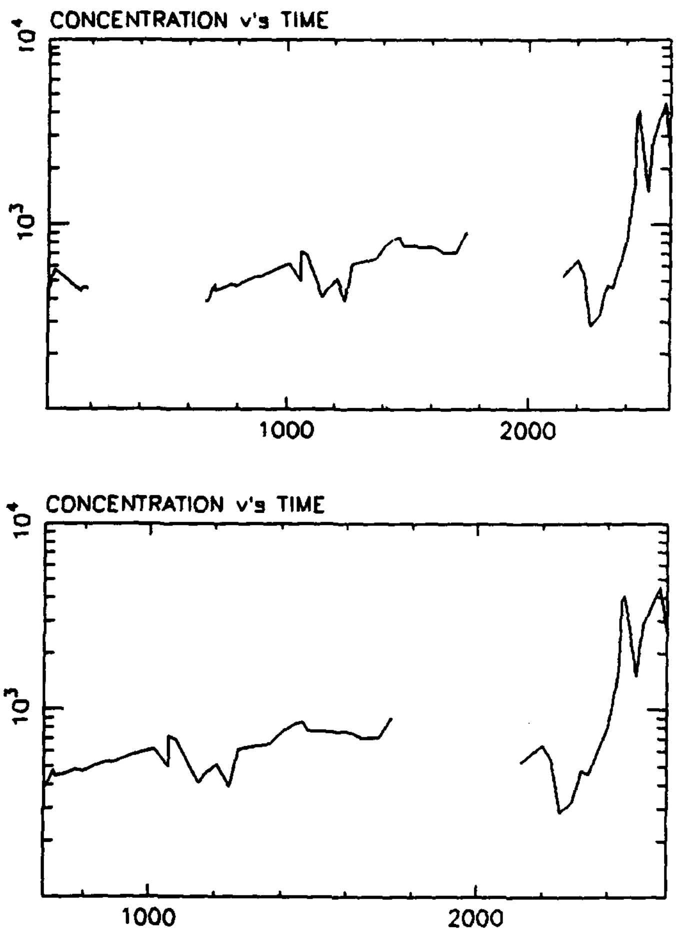


Fig. 29 The variation of nucleus concentration as a function of time (shown here in minutes) for the June 1 - June 3 period at Mace Head. The upper curve covers all of the measurement period.

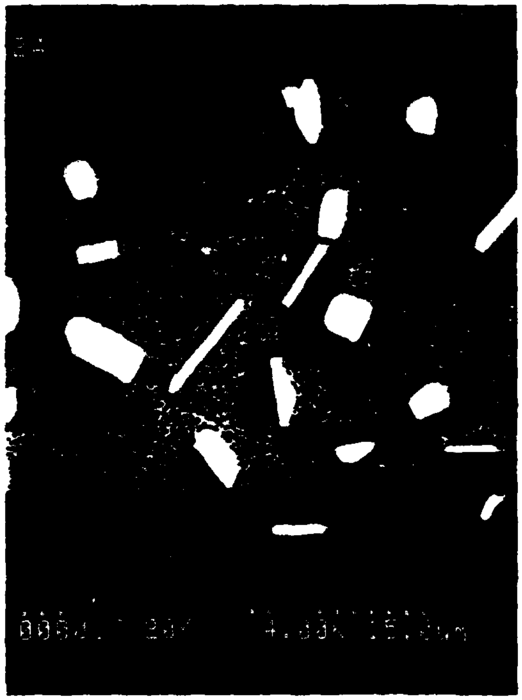


Fig. 30 Scanning electronmicrograp of ambient aerosol samples at Mace Head and collected on a nuclepore filter of pore size 0.2 μm.

Upper curve: Aerosol sampled on April 20 1988 with a SW air mass (oceanic).

Lower curve: Aerosol sampled on April 22 1988 with a E air mass (continental).

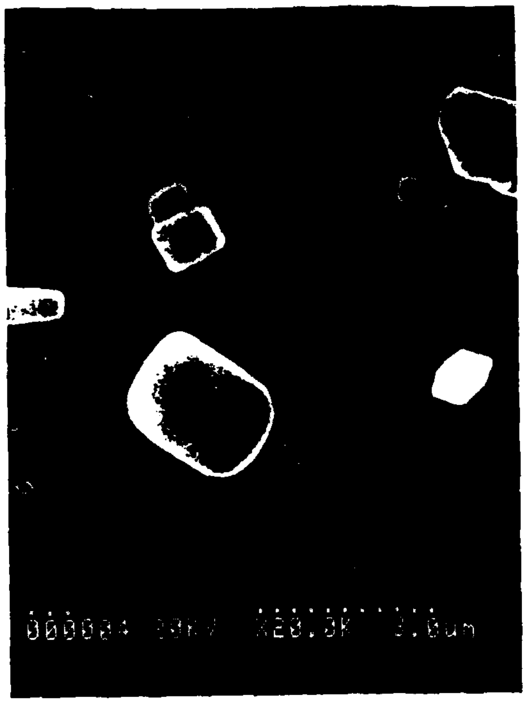


Fig. 31   As per Fig. 30 except for:

Upper curve:

Period June 2 1988 with a SW air mass.

Lower curve :

Period June 2 1988 with a SW air mass.

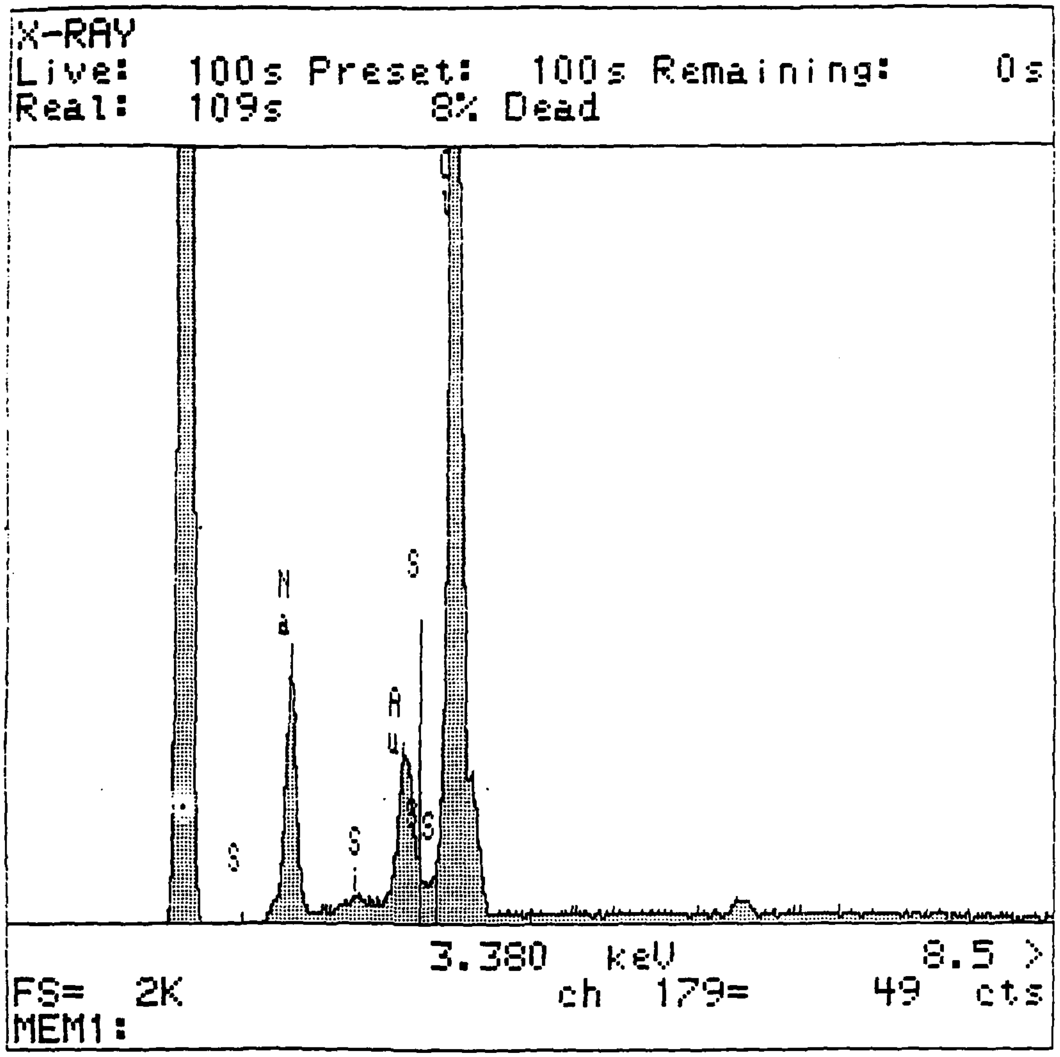


Fig. 32 X-ray Energy Dispersive Analysis of Elements at Mace Head. Peaks for Chloride, Sodium and Sulphur are evident. The Au peak (filter coating) should be disregarded.

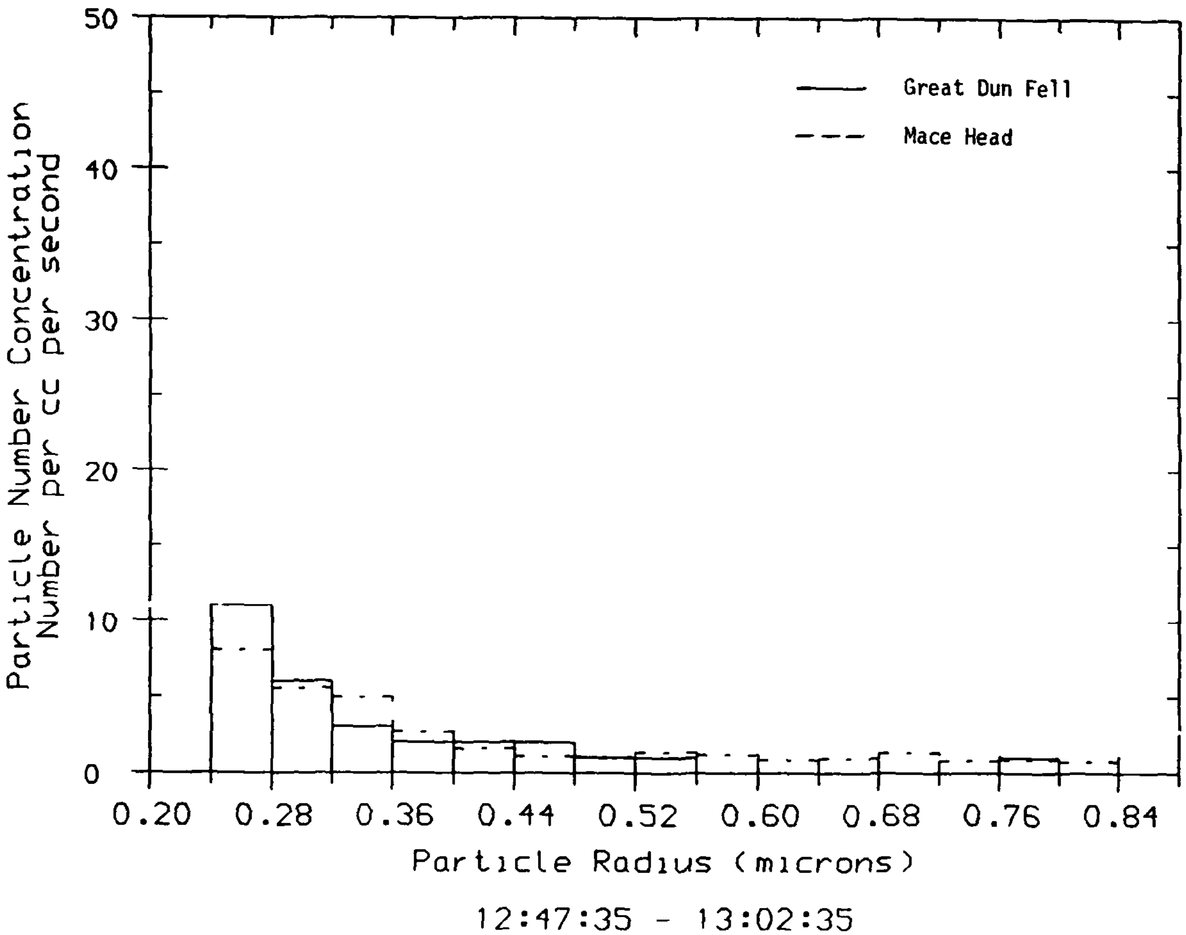
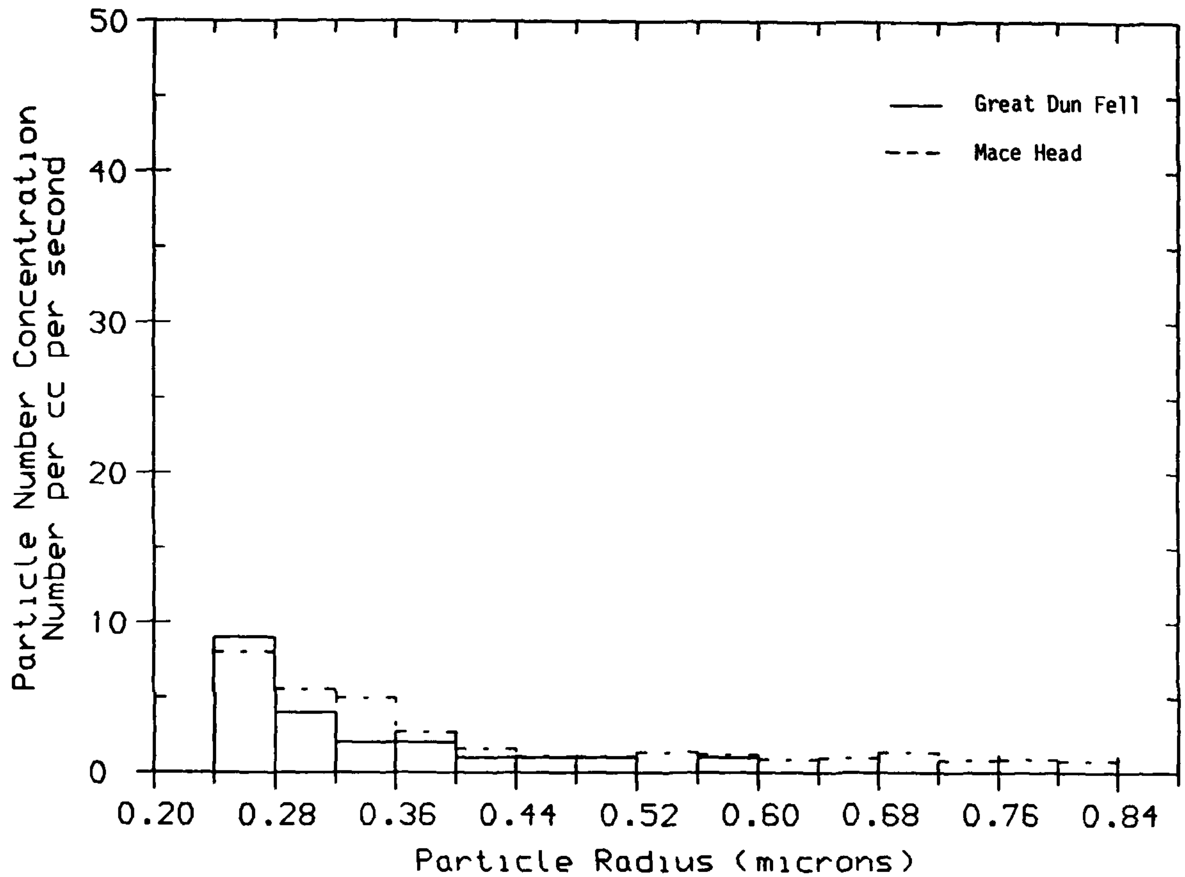


Fig. 33 A comparison of particle number concentration measured at Mace Head



13:02:36 - 13:17:36